THE
PHYSICAL REVIEW
A Journal of Experimental and Theoretical Physics Established by E. L. Nichols in 1893
VOL. 57, No. 12 JUNE 15, 1940 SECOND SERIES

Collisions of Alpha-Particles with Carbon Nuclei*

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(Received April 1, 1940)

More than 750,000 thorium (C+C′) alpha-particle cloud tracks have been photographed with a stereoscopic camera using a cloud chamber filled in one series with methyl-chloride and helium, and in another with acetylene and helium. For each series a range-velocity curve for carbon has been constructed by plotting the measured ranges, reduced to standard air values, against the calculated velocities for a total of 55 selected alpha-particle-carbon collisions. These curves are used for testing the validity of the method of reducing the ranges of heavy recoil nuclei in any gas to standard air values. Partial validity of the method has been observed in the present instance. An empirical relationship between the total mean straggling (in range), the atomic numbers of the atoms comprising the absorbing medium, and the atomic number of the recoil nucleus is presented. A brief analysis of the effectiveness of certain errors on the range-velocity results is given.

INTRODUCTION

IN 1923 a method of analysis of forked cloud tracks was published by Blackett[1] who calculated range and velocity values for recoil oxygen, hydrogen, and helium nuclei. The work was considerably extended and refined by Blackett and Lees[2] who added relations for nitrogen and argon to those already mentioned. Feather[3] has obtained range and velocity values for fluorine nuclei from a cloud-chamber study of the scattering of alpha-particles in carbon tetrafluoride. He investigated at the same time the cloud tracks of eight recoil nuclei supposed to be carbon. The range-velocity relation for neon has been investigated by Eaton,[4] for sulphur by Anthony[5] and for deuterium and neon by McCarthy.[6]

The current practice in the presentation of the above information has been to reduce all range measurements to the values they would have had in dry air at 15°C and at normal atmospheric pressure, regardless of the nature of the gas in which the measurements have been made. The classical method of obtaining this reduction depends upon assumptions to which objection may be made, and has been used only because no other satisfactory method of reducing the results to this standard has been presented. One of the objects—important in itself—of the present paper is to test the validity of this procedure. Such an investigation must at present be purely

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[1] P. M. S. Blackett, Proc. Roy. Soc. **A103**, 62–78 (1923).
[2] P. M. S. Blackett and D. S. Lees, Proc. Roy. Soc. **A134**, 658–671 (1932).
[3] N. Feather, Proc. Roy. Soc. **A141**, 194–209 (1933).
[4] W. W. Eaton, Phys. Rev. **48**, 921–928 (1935).
[5] R. L. Anthony, Phys. Rev. **50**, 726–732 (1936).
[6] J. T. McCarthy, Phys. Rev. **53**, 30–34 (1938).

experimental in nature, since the dependence of the effective charge of a moving atomic nucleus on its kinetic energy cannot be obtained without experiment.

Among the lighter elements carbon has received but little attention, presumably because there appears to be small chance of observing any disintegration effects in the cloud chamber. Hence material for a range-velocity curve for carbon is still required. In part for this reason, but to a greater extent because carbon represents the best available material for accurate tests on the classical method of range reduction, it was chosen for the present work.

The range-velocity curves which have been obtained so far can be divided into two groups: one for which the method of reduction of the ranges of heavy recoil nuclei to standard air conditions is known to be reasonably valid, and the other for which the method contains an unproved assumption. The first group includes nitrogen and oxygen, as either alone approximates air in composition. Helium is included, as the reduction to standard air conditions is made by measuring the stopping power of the gas for alpha-particles. Hydrogen and deuterium are also included since experimental measurements[7] indicate that the relative atomic stopping power of a gas is the same for protons and deuterons as for alpha-particles. The second group contains fluorine, neon, argon, sulfur, and the elements studied in the present investigation, carbon and chlorine.

The method of reduction of a set of forked tracks to standard air conditions can be described as a "demagnification" of the recombination photograph of the tracks to the size where undeviated alpha-particle tracks have the length they would have had in standard air, suitable adjustments being made, if necessary, for variation in the demagnification factor (relative stopping power) with the range of the alpha-particles. The objection to this procedure is that there is no apparent reason to believe that the demagnification factor is the same for a heavy recoil nucleus as for an alpha-particle when a transfer from one gas to another is involved in standardizing the results. To provide an answer to this objection, data on the range and velocity of carbon recoil nuclei were obtained from two distinctly different gas mixtures and are compared in this paper.

[7] C. Gerthsen, Ann. d. Physik [5] **5**, 657–669 (1930).

Theory

One of the outstanding advantages in making a cloud chamber analysis of the range and velocity of atomic nuclei is that the physical laws involved in the calculations, namely the conservation of energy and of linear momentum, are most fundamental and well established.[8] It is assumed that all of the forked tracks studied in the present paper represent nuclear collisions which are elastic* in nature. Hence Eqs. (1), (2) and (3), which follow from the laws of conservation of energy and of linear momentum, are employed in the present analysis to obtain values of the initial velocities of recoil nuclei:

$$\frac{m}{M}=\frac{\sin\Phi}{\sin(2\theta+\Phi)}, \tag{1}$$

$$u=\frac{\sin(2\theta+\Phi)}{\sin\theta}v, \tag{2}$$

$$u=\frac{2V\cos\theta}{\{1+m/M\}}. \tag{3}$$

Here M and m represent the masses of the alpha-particle and colliding nucleus, respectively. V represents the velocity of the alpha-particle just before collision, while v and u represent the respective velocities of alpha-particle and nucleus just after collision. Φ is the angle of deflection of the alpha-particle, while θ represents the angle made by the trajectory of the struck nucleus and the original, or forward, direction of the incident alpha-particle. For convenience, Φ and θ are both taken to be positive when they lie on opposite sides of the direction of the incident alpha-particle.

Experimental Procedure

The cloud chamber used in the present study is of the conventional sylphon type, following

[8] F. Joliot, Comptes rendus **192**, 1105–1107 (1931).

* Calculation of distances of closest approach in the collisions investigated here show that only Coulomb field scattering was effective.

closely the design published by Dahl, Hafstad and Tuve.[9] It has the outstanding advantages when used for measurements on the cloud tracks of recoil nuclei of having a large diameter and of being easily adjusted for work over a wide range of pressures. The stereoscopic camera used in conjunction with the cloud chamber has previously been described.[10]

For the present work, the cloud chamber was altered in several respects from its original form. Two aluminum wedge-shaped latches which automatically lock the cloud chamber piston at the lowest point of its expansion stroke have been installed, as well as a control system regulating the air pressure producing the cloud chamber expansion, so that all expansions were of the same magnitude. Cloud track illumination was provided by two banks of spotlights, each containing eight fifty-candlepower tungsten filament lamps arranged in such a way that light was scattered uniformly by all parts of each track to each mirror into the stereoscopic camera.

A wedge-shaped button of polished silicon steel was used as an alpha-particle source. Powdered material containing thorium was placed on a conducting plane in a closed container. Above this the wedge source was placed and maintained at a negative potential of about 300 volts relative to the plane. Due to the geometry of the wedge and plane the electric field has a maximum gradient along the sharp edge of the wedge, so that the greatest density of recoil thorium A ions is found in this region, effectively forming a strong line source of Th (C+C′) alpha-particles.

Cloud tracks were photographed on Agfa ultra-speed panchromatic film which is efficient under tungsten lamp illumination. D-76 fine-grain developer was used. In recombining track images, the film was replaced in the stereoscopic camera in the same position it had when the cloud chamber was photographed, and the real images of the stereoscopic pair of negatives were reprojected onto a screen placed below a thickness of glass equivalent to that in the top of the cloud chamber. If the film has been replaced correctly, the recombined images of a given track will have the same dimensions on the screen that the original track had in the cloud chamber. Forked tracks which are coplanar can be recombined in this manner by suitable orientation of the combining screen, which has one linear and two angular adjustments.

Reprojection recombinations were made directly onto the sensitive print paper by inserting a ruby light filter in the reprojection system, a flicker method of alternating the images on the screen being used. The ruby light filter was simply withdrawn when it was desired to make a reprojection print. In general the two components of a given stereoscopic combination were registered on separate prints, Eastman Kodaline extreme contrast paper being used.

Survey of Errors

The main factors which influenced the accuracy of the experimental results are as follows.

1. Limits of accuracy imposed by the apparatus

A study of the geometry of the alpha-particle source and of the characteristics of the cloud-chamber expansion stroke indicated that, due to an induced straggling effect, the measurement of the relative stopping power has been indefinite to the extent of about ±6 percent, in addition to statistical straggling effects, in the work done by Anthony and McCarthy. It has also been estimated that corrective alterations have reduced this extra straggling effect to about ±1 percent in the present work.

While obtaining a precise measurement of the relative stopping power is ideal in the type of analysis presented here, it is worthy of note that even an approximate measurement of this quantity yields relatively accurate results on a range-velocity curve. In support of this statement, it is found by inspection that practically all the alpha-particle spurs involved in the analysis have standard air ranges which are between 1.0 and 3.0 cm in length. Graphical differentiation of the standard range-velocity curve for alpha-particles shows that the ratio of the fractional errors in measured values of alpha-particle velocity U and range R, that is:

$$f_U/f_R=(R/U)(dU/dR) \qquad (4)$$

[9] O. Dahl, L. R. Hafstad and M. A. Tuve, Rev. Sci. Inst. **4**, 373–378 (1933).
[10] F. N. D. Kurie, Rev. Sci. Inst. **3**, 655–667 (1932).

is nearly constant for this region of range values, having the value 0.40 ± 0.06. It is understood here that either U or R is a measured value from which the other is obtained by reference to the standard range-velocity curve.

If we suppose that the measured value of the relative stopping power is N (percent) too large, due to induced range-straggling, then the measured standard air length r of the track caused by the heavy recoil nucleus will also be N too large. By Eq. (4), the recoil velocity of the alpha-particle involved in the collision will be $0.4\,N$ too large due to this cause, since its standard air range will be N too large. As u is a linear function of v or V by Eq. (2) or (3), it will be $0.4\,N$ too large due to the induced range-straggling error, as U can represent either velocity v or V. Hence the range and velocity errors for the recoil nucleus tend to cancel each other to an important degree in the method of analysis employed here.

As errors in the measurement of the relative stopping power follow a Gaussian distribution function, the probability of making a 6 percent error in the work of Anthony or of McCarthy would be small. These considerations indicate that the range-velocity curves presented by these workers can be considered to be reasonably reliable, insofar as the error due to induced straggling is concerned.

To study the accuracy of the stereoscopic camera in photography and reprojection, a flat white card on which a set of concentric circles and radial lines were drawn was photographed in positions representative of all possible forked tracks, and the recombination prints were compared with the original card. From these it was found that:

(a) The images registered on the print paper were 0.8 percent smaller than the original object, probably due in part to shrinkage of the photographic emulsions involved.

(b) The fidelity of reprojection of the pattern was greatly improved by storing the exposed and developed film prior to reprojection in rolls with the emulsion outside rather than inside.

(c) An impartial assignment of the relative weight of pairs of prints in various orientations was obtained from the ratio of the mean error in length and angle measurements in the corresponding orientations in the test series.

2. Limits of accuracy imposed by observational errors

In order to estimate the magnitudes of this type of error, two completely separate analyses were carried out on a number of the collision tracks involving chlorine. Comparison of these two analyses yielded the following information:

Observable	Φ	S	r
Mean variation in measurement, percent	± 0.8	± 1.0	± 3.0

where S represents the relative stopping power of the cloud chamber gas. Recoil spur length r was measured by an eyepiece micrometer (fixed scale type). The average fractional error in measuring r for chlorine nuclei will be greater than for carbon, since the latter produces longer recoil spurs on the average.

3. Limits of accuracy imposed by indefiniteness in auxiliary data used

As no experimental information on the variation of the relative atomic stopping power of chlorine for alpha-particles is available, this relation was interpolated from Mano's[11] calculations. The magnitude and variation of the stopping power of carbon was similarly obtained, and was found to agree reasonably well with the experimental relation published by Livingston and Bethe,[12] thus providing a check on the validity of the interpolation method.

With this information a gas and vapor mixture, containing in volume percent, 52 CH_3Cl, 26 He, 16 C_2H_5OH, 6 H_2O, was employed, the relative atomic stopping power of which should vary with velocity by less than 2 percent over the range of alpha-particle velocities used. In agreement with this calculation, it was found that the stopping power of this mixture had the same value when calculated from measured values of the ranges of Th C and Th C′ alpha-particle groups in the gas. Hence no correction for variation in stopping power of the gas with alpha-particle velocity was felt to be necessary.

[11] G. Mano, J. de phys. et rad. [7] **5**, 628–634 (1934).
[12] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 246–390 (1937).

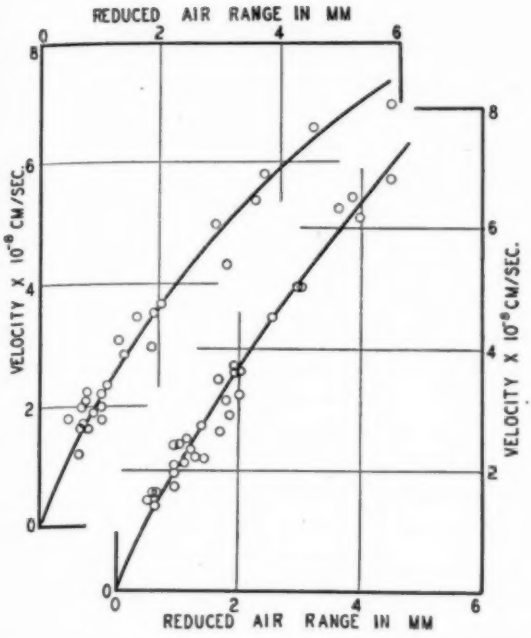

FIG. 1. Range-velocity curve for carbon recoil atoms in methyl-chloride and helium (upper curve) and in acetylene and helium (lower curve).

In the work with acetylene the following gas mixture was used: 49 C_2H_2, 40 He, 8 C_2H_5OH, 3 H_2O. It was necessary in this case to make correction for the variation in stopping power with velocity. This was calculated from experimental data[12, 13] and checked against the variation in stopping power values as found from measuring the ranges of the Th C and Th C′ alpha-particle groups in the gas. These were found to agree reasonably well, and hence an average value of the two variations was employed.

RESULTS

The cloud tracks of approximately 750,000 alpha-particles have been photographed in about 12,000 exposures. Of these, approximately two-thirds are caused by the long-range Th C′ alpha-particles, the remainder being due to Th C. In Fig. 1 are shown range-velocity curves for carbon from methyl-chloride and from acetylene, the experimental points of which have been obtained by the general method previously outlined.

The relatively few forked tracks used in constructing these curves have been selected from a much larger number available, the criterion for selection being that all or most parts of a collision fork must be clearly visible. In the work with methyl-chloride a considerably larger number of experimental points for chlorine than for carbon were obtained. This is to be expected when the alpha-particles do not approach nearer to the nucleus than distances at which only Coulomb fields of force are effective, for then the collision cross section for chlorine will be $\{17/6\}^2 \doteq 8$ times as large as that for carbon nuclei.

However, as a much larger fraction of the kinetic energy of an alpha-particle can be transferred to a carbon nucleus than to one of chlorine, a larger fraction of the collisions with carbon will produce measurable spurs, than in the case of chlorine, as was found to be the case in this work.

DISCUSSION AND CONCLUSIONS

The degree of equivalence between the range-velocity curves for recoil carbon nuclei in methyl-chloride-helium and acetylene-helium gas mixtures is seen by comparison of the curves in Fig. 1. There appears to be a definite coincidence in the location of the two curves only for standard air ranges of less than 3 mm, beyond which point there is an increasing divergence. No conclusion of general applicability can be drawn from this divergence concerning the degree of validity of the method used in reducing ranges of heavy recoil nuclei to standard air values from measurements in gases other than air. However, in the case of the carbon-containing gases used here, and probably in others, the use of the current method of applying range-velocity data obtained in one gas mixture to collisions in another gas will induce a characteristic error when high energy recoil nuclei are being studied.

It was found that the range-velocity curves for carbon, obtained from those already published for nitrogen and for alpha-particles by use of Blackett and Lees[2] relation $r \propto mZ^{-\frac{1}{2}}f(u)$, where m is the mass of the recoil nucleus and Z its atomic number, disagreed both in shape and in placement with the curves presented in this paper. Feather's[3] experimental curve for recoil nuclei believed to be carbon, very nearly coincides with the upper part of Fig. 1, falling between the two curves for carbon.

[13] R. W. Gurney, Proc. Roy. Soc. **A107**, 340–349 (1925).

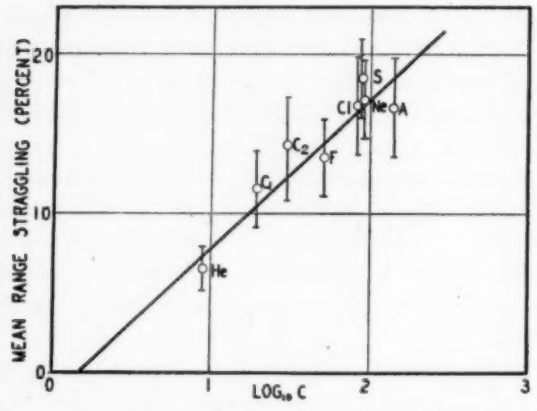

FIG. 2. Empirical relation between atomic numbers of recoil atom and absorbing medium, and the average range straggling of experimental points.

It has been found that the fractional error involved in the measurement of cloud track lengths is of the order of ± 3 percent for short tracks, and the probable error in the measurement of the recoil velocity u is approximately ± 2 percent. Yet measurement shows that the average total range-straggling of experimental points from the experimental curve has a value of more than 10 percent for each of the range-velocity curves of heavy recoil nuclei for which a sufficient number of experimental points have been obtained with which to make an analysis.

It is customary to attribute the additional range-straggling to statistical variations in the interaction of the recoil nucleus and its remaining orbital electrons with the atoms of the medium through which the nucleus passes. If the interaction is entirely between orbital electrons, the degree of straggling should be a function of the product of the effective nuclear charge of the recoil atom (equal to the number of removed electrons) with the effective atomic number* B of the absorbing atoms. If, however, there is a considerable probability of slight direct interactions of the recoil nucleus with nuclei of the absorbing medium, the amount of straggling would be a function of the product (C) of the atomic number of the recoil atom with B.

* If the medium is composed of atoms having atomic numbers $A_1, A_2, \cdots A_n$, and relative abundances $f_1, f_2, \cdots f_n$, respectively, the effective atomic number of the atoms in the medium is taken here to be

$$B=\sum_{s=1}^{s=n} A_s f_s \quad \text{where} \quad \sum_{s=1}^{s=n} f_s = 1.$$

In order to investigate the possibilities of such relations, the average of the total fractional range-straggling for all ranges having standard air values between 0.5 and 2.0 mm was measured for each of the published range-velocity curves for He, F, Ne, S, A, Cl, and the two curves for carbon presented in this paper. Equivalent range sections of each range-velocity curve were chosen so that the average number of absorbing atoms encountered by a recoil nucleus would be the same for all nuclei. The section of the curves selected was that which was most richly populated with experimental points.

It proved impossible with the very rough information available to indicate what the predominating cause of straggling among the heavy recoil nuclei might be. However, an interesting empirical relation between the product C and the total range straggling was obtained and is shown in Fig. 2. It indicates that the practice of mixing a heavy gas with a light one merely to increase the relative stopping power of the latter[6] is a less desirable condition than using the lighter gas at a pressure sufficiently great to give a relative stopping power equivalent to that of the mixture.

In addition to many examples of secondary branches and deflections in the tracks of recoil nuclei published by other authors, notably those of Joliot,[14] a series of such branched tracks has been observed by the writer. These indicate that a considerable nuclear collision cross section exists for heavy recoil atomic nuclei in a medium containing heavy atomic nuclei. Estimation of the expected frequency of slight collisions between such heavy nuclei, from observation of the distribution of points on range-velocity curves and also from the theory of Coulomb field scattering, indicates that such encounters occur with a frequency such that they cannot be neglected in an explanation of the large straggling effect noted above.

It is a pleasure for the writer to thank Professor A. F. Kovarik for his continued advice and encouragement throughout the course of this work. He wishes also to acknowledge the valuable assistance given by Mr. W. L. Davidson, Jr., in checking calculations.

[14] F. Joliot, J. de phys. et rad. [7] **5**, 219–225 (1934).

Heavy Electron Pair Theory of Nuclear Forces

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(Received April 1, 1940)

The interpretation of the proton-proton scattering experiments on the basis of Bethe's neutral meson theory leads to a range of nuclear forces $\approx \frac{1}{2}\hbar/\mu c$ (μ is meson mass). This implies a mass for the field particle equal to twice the mass of the cosmic-ray meson. It has also been found that a meson theory of nuclear forces, involving the emission and absorption of single charged mesons obeying Bose statistics, does not give the right sign of the quadrupole moment of the deuteron. In view of these difficulties and the fact that the single meson theory cannot correctly explain the β-decay anyway, we have investigated the consequences of a heavy electron pair theory of nuclear forces. The heavy electrons are assumed to be identical with electrons in every respect ("hole" theory, Fermi statistics, etc.) except that their rest mass is taken equal to the cosmic-ray meson mass. A tensor interaction between the nuclear particles and the heavy electron pair field can alone account for the spin dependence of the nuclear forces and the positive quadrupole moment of the deuteron. It is interesting that a pseudo-vector interaction gives both the wrong sign of the quadrupole moment and too much repulsion. The potential function between two nuclear particles behaves at large distances, r, as $e^{-2kr}/r^{2.5}$, $k \approx \mu c/\hbar$ so that the range is effectively one-half the single meson range, and is directly connected with the rest mass of the heavy electron pair field (in contrast to the Gamow-Teller pair theory). At small r, the potential goes as $1/r^5$ so that one has to cut off in the same way as in the original electron-neutrino theory. The advantage of the heavy electron pair theory over a neutral meson theory is that it deals with particles which can be identified with the cosmic-ray mesons.

§1. INTRODUCTION

IN a recent paper[1] Bethe has examined quantitatively the "symmetrical" and "neutral" formulations of the single meson field theory of nuclear forces. He finds that Kemmer's "symmetrical" theory[2] (according to which both charged and neutral mesons obeying Bose statistics are emitted by nuclear particles) leads to several serious difficulties. First, the "cut-off" distance must be considerably larger than the natural range of nuclear forces following from the finite mass of the field particle. This implies that the nuclear force exists only at distances greater than 3×10^{-13} cm and is in conflict with all the strong evidence for short range forces such as the large binding energies of the triton and α-particle as compared with the deuteron, the almost total absence of p wave scattering of protons by protons at low energies, etc. Secondly, the quadrupole moment of the deuteron turns out to have the wrong sign and to be much too large. Finally any attempt to account for the β-decay by means of the "symmetrical" theory predicts a lifetime of the meson itself which is much shorter than that permitted by the cosmic-ray data. In addition to the shortcomings of the "symmetrical" theory revealed by Bethe's calculations, attention has also been called by Weisskopf[3] to the "peculiar" character of the internal charge distribution of an elementary particle with Bose statistics (scalar or vector meson) in contrast to the "well-behaved" charge distribution of an elementary particle obeying Fermi statistics and with spin one-half (electron).

On the other hand, Bethe finds that the assumption of an interaction of the nuclear particles with a "neutral" meson field alone leads to both a reasonable "cut-off" distance and the right sign and magnitude of the quadrupole moment of the deuteron. The neutral mesons are assigned Bose statistics, spin one and the mass experimentally found (150–200 electron masses) for the penetrating component of the cosmic rays. However, investigations by Breit and his collaborators[4–6] on the theoretical interpretation of the experimental data on proton-proton scattering and neutron-proton scattering, using Bethe's form of "neutral" meson theory, point

[1] H. A. Bethe, Part I, Phys. Rev. **57**, 260 (1940); Part II, **57**, 390 (1940).
[2] N. Kemmer, Proc. Camb. Phil. Soc. **34**, 354 (1938).
[3] V. Weisskopf, Phys. Rev. **57**, 1066A (1940).
[4] L. E. Hoisington, S. S. Share and G. Breit, Phys. Rev. **56**, 884 (1939).
[5] G. Breit, C. Kittel and H. M. Thaxton, Phys. Rev. **57**, 255 (1940).
[6] C. Kittel and G. Breit, Phys. Rev. **56**, 744 (1939).

to a range of nuclear forces of about $\frac{1}{2}\hbar/\mu c$ (μ is meson mass). This implies a mass of the field particle equal to about twice the mass of the cosmic-ray meson. Thus far no real evidence exists for neutral mesons of either the same mass as the charged mesons or twice their mass; moreover it would seem desirable to have recourse only as a last resort to a theory which depends on the postulation of unobserved particles.

In view of the above objections to Bethe's "neutral" theory, we have explored the possibility of a theory which only makes use of particles which can be identified with the observed cosmic-ray mesons, leads to a range of nuclear forces of $\frac{1}{2}\hbar/\mu c$, and at the same time retains the following desirable features of Bethe's "neutral" theory:

(a) Single force hypothesis; only one type of interaction between the nuclear particles and the field is needed to give the relative positions of the singlet and triplet levels of the deuteron and the sign of the quadrupole moment.

(b) Like and unlike particle forces arise in the same order of perturbation theory.

(c) Only a small admixture of D function is required in the deuteron ground state to give the magnitude of the quadrupole moment and therefore does not contradict the "almost" additivity of the magnetic moments of the neutron and proton.[7]

(d) A steep increase of the potential at small distances with the consequent possibility of accounting for the larger binding energies of the triton and α-particle.

§2. Interaction of Nuclear Particles with Heavy Electron Pair Field

We assume that the field consists of charged particles which obey Fermi statistics and possess a rest mass equal to the cosmic-ray meson mass. It is also assumed that the Dirac equation, implying a spin one-half, holds for these particles and that "hole" theory can be used to explain the presence of both signs of charge. In other words, the field particles are really *heavy electrons* —identical with electrons in every respect except for their mass which is taken to be about 200 times the electron mass. In this paper we reserve the term "meson" for the particles observed in cosmic-ray experiments and refer to the field particles of negative charge as heavy electrons and to those of positive charge as heavy positrons.

As the Hamiltonian which describes the interaction between the nuclear particles (neutron or proton) and the heavy electron-positron field, we first consider the most general type: a linear combination of the five possible relativistic invariants[8] which can be used when no derivatives of the wave functions are involved. Later we shall adopt Bethe's "single-force hypothesis" and set all constants except one equal to zero. Since we work in the non-relativistic approximation for the nuclear particles, all terms involving the Dirac α's of the nuclear particles can be neglected and the Dirac β's of the nuclear particles can be set equal to one. Thus the pseudo-scalar interaction disappears entirely and we get:[9]

$$H=G\{C_1\Psi^*\Psi\psi\beta\psi+C_2\Psi^*\Psi\psi^*\psi + C_3\Psi^*\boldsymbol{\sigma}_n\Psi\psi^*\beta\boldsymbol{\sigma}_e\psi+C_4\Psi^*\boldsymbol{\sigma}_n\Psi\psi^*\boldsymbol{\sigma}_e\psi\}, \quad (1)$$

where Ψ is the quantized Pauli two-component wave function, $\boldsymbol{\sigma}_n$ the two-component spin operator for the nuclear particle; ψ is the quantized Dirac wave function, $\boldsymbol{\sigma}_e$ the four-component spin operator for the heavy electron. The asterisk signifies creation and its absence destruction; the destruction of a negative-energy heavy electron is equivalent to the creation of a heavy positron in accordance with the Dirac "hole" theory. The second-order perturbation energy which describes the transition of a proton in a specified state π and a neutron in a specified state ν to two other specified states π', ν' is given by:

$$W=-\sum_b \frac{H_{bd}{}^*H_{ab}}{E_b-E_a}-\sum_c \frac{H_{cd}H_{ac}{}^*}{E_c-E_a}, \quad (2)$$

where a and d refer to the initial and final composite states, respectively, and b and c to the

[7] L. W. Alvarez and F. Bloch, Phys. Rev. **57**, 111 (1940).

[8] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 82 (1936).

[9] The same coupling constants are taken for both neutron and proton because of the equality of like and unlike particle forces; consequently a calculation performed for proton-neutron is just as valid for proton-proton or neutron-neutron (cf. below).

two intermediate states with a heavy electron pair present in the field—the pair being emitted in the first case by a proton and in the second case by a neutron. In the transition to the final state the pair is reabsorbed by the neutron or the proton. Since these two possibilities are completely equivalent we can just as well consider one and multiply the result by two; we consider the first. We get for the perturbation energy in the transition $\pi,\ \nu\rightarrow\pi',\ \nu'$

$$W=-2G^2\sum_{ik,=1}^{4}C_iC_k\sum_b\left[\frac{1}{(p^2+1)^{\frac{1}{2}}+(q^2+1)^{\frac{1}{2}}}\int\int(\Psi_{\nu'}{}^*O^i\Psi_\nu)(\psi_q{}^*O^i\psi_p)(\psi_p{}^*O^k\psi_q)(\Psi_{\pi'}{}^*O^k\Psi_\pi)d\tau_1d\tau_2\right], \quad (3)$$

where O^i, O^k are for the nuclear particles I (identity matrix) or $\boldsymbol{\sigma}_n$, and for the field particles β, I, $\beta\boldsymbol{\sigma}_e$ or $\boldsymbol{\sigma}_e$; p always refers to the momentum of a positive energy heavy electron and q to that of a negative energy heavy electron. Since our unit of momentum is μc, of energy μc^2, of length $\hbar/\mu c$ (μ is heavy electron mass), $(p^2+1)^{\frac{1}{2}}$ and $(q^2+1)^{\frac{1}{2}}$ represent energies of the heavy electrons in the intermediate state. Moreover the recoil energy of the nuclear particle has been neglected since its rest mass is large. Hence, if plane waves are taken for the heavy electrons, the interaction potential in configuration space becomes:

$$J_{NP}(\mathbf{r})=\sum_{i,k=1}^{4}C_iC_kO^iO^kJ_{ik}$$

with

$$J_{ik}=-2G^2\int d\mathbf{p}\int d\mathbf{q}\frac{\exp\left[i(\mathbf{q}-\mathbf{p})\cdot\mathbf{r}\right]}{(p^2+1)^{\frac{1}{2}}+(q^2+1)^{\frac{1}{2}}}\sum_{\text{spins}}{}'\,(\bar{\psi}_q{}^*O^i\bar{\psi}_p\bar{\psi}_p{}^*O^k\bar{\psi}_q), \quad (4)$$

where $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2$, $\mathbf{r}_1$ specifies the position of one nuclear particle and $\mathbf{r}_2$ that of the other, and the $\bar{\psi}$'s are the amplitudes of the plane waves. The integration is over all possible values and directions of the momentum and the summation over both orientations of spin corresponding to the positive energies of one heavy electron and the negative energies of the other. To evaluate the spin summation we insert operators before the $\bar{\psi}$'s so that we may sum over both signs of the energy for both heavy electrons. Thus:

$$\frac{1}{2}\left(1+\frac{\boldsymbol{\alpha}\cdot\mathbf{p}+\beta}{(p^2+1)^{\frac{1}{2}}}\right)\bar{\psi}_p=\bar{\psi}_p \quad \text{for positive energies}$$
$$=O \quad \text{for negative energies,}$$

$$\frac{1}{2}\left(1-\frac{\boldsymbol{\alpha}\cdot\mathbf{q}+\beta}{(q^2+1)^{\frac{1}{2}}}\right)\bar{\psi}_q=\bar{\psi}_q \quad \text{for negative energies}$$
$$=O \quad \text{for positive energies}$$

and we get:

$$P^{ik}=\sum_{\text{spins}}{}'\bar{\psi}_q{}^*O^i\bar{\psi}_p\bar{\psi}_q{}^*O^k\bar{\psi}_q$$
$$=\sum\tfrac{1}{4}\bar{\psi}_q{}^*O^i\left(1+\frac{\boldsymbol{\alpha}\cdot\mathbf{p}+\beta}{(p^2+1)^{\frac{1}{2}}}\right)\bar{\psi}_p\bar{\psi}_p{}^*O^k\left(1-\frac{\boldsymbol{\alpha}\cdot\mathbf{q}+\beta}{(q^2+1)^{\frac{1}{2}}}\right)\bar{\psi}_q$$

(over spins for both signs of energy)

$$=\sum\tfrac{1}{4}\bar{\psi}_q{}^*\left[O^i\left(1+\frac{\boldsymbol{\alpha}\cdot\mathbf{p}+\beta}{(p^2+1)^{\frac{1}{2}}}\right)O^k\left(1-\frac{\boldsymbol{\alpha}\cdot\mathbf{q}+\beta}{(q^2+1)^{\frac{1}{2}}}\right)\right]\bar{\psi}_q$$

or

$$P^{ik}=\tfrac{1}{4}\,\text{Spur}\left[O^i\left(1+\frac{\boldsymbol{\alpha}\cdot\mathbf{p}+\beta}{(p^2+1)^{\frac{1}{2}}}\right)O^k\left(1-\frac{\boldsymbol{\alpha}\cdot\mathbf{q}+\beta}{(q^2+1)^{\frac{1}{2}}}\right)\right]. \quad (5)$$

P^{ik} can be found for different choices of O^i and O^k by means of the usual rules for evaluating the spurs of products of Dirac operators. The cross terms $(i \neq k)$ do not give any contribution when integrated over momentum space and so we get for the interaction energy between a neutron and a proton:

$$J_{NP}(r) = -2G^2 \int d\mathbf{p} \int d\mathbf{q} \exp\left[i(\mathbf{q}-\mathbf{p})\cdot\mathbf{r}\right] \cdot \frac{1}{(p^2+1)^{\frac{1}{2}}+(q^2+1)^{\frac{1}{2}}} \Bigg\{ (C_1^2+C_2^2)\left(1-\frac{1}{(p^2+1)^{\frac{1}{2}}(q^2+1)^{\frac{1}{2}}}\right)$$
$$+(C_1^2-C_2^2)\frac{\mathbf{p}\cdot\mathbf{q}}{(p^2+1)^{\frac{1}{2}}(q^2+1)^{\frac{1}{2}}}+(C_3^2+C_4^2)\left[\sum_{r,s=1}^{3}\sigma_r^N\sigma_s^P\delta_{rs}\left(1-\frac{1}{(p^2+1)^{\frac{1}{2}}(q^2+1)^{\frac{1}{2}}}\right)\right]$$
$$+(C_3^2-C_4^2)\left[\sum_{r,s=1}^{3}\sigma_r^N\sigma_s^P\left(\frac{p_rq_s+p_sq_r}{(p^2+1)^{\frac{1}{2}}(q^2+1)^{\frac{1}{2}}}-\frac{\delta_{rs}\mathbf{p}\cdot\mathbf{q}}{(p^2+1)^{\frac{1}{2}}(q^2+1)^{\frac{1}{2}}}\right)\right]\Bigg\}. \quad (6)$$

After the angular integrations are performed we get the following two characteristic integrals:

(A) $$\int_0^\infty pdp\int_0^\infty qdq\frac{\sin pr\sin qr}{(p^2+1)^{\frac{1}{2}}+(q^2+1)^{\frac{1}{2}}}=\frac{\pi}{2}\left(-\frac{\partial}{\partial z}\right)\int_1^\infty dq(q^2-1)^{\frac{1}{2}}e^{-qz}=-\frac{\pi}{2}\frac{\partial}{\partial z}\left[\frac{K_1(z)}{z}\right],$$

(B) $$\int_0^\infty pdp\int_0^\infty qdq\frac{\sin pr\sin qr}{(p^2+1)^{\frac{1}{2}}+(q^2+1)^{\frac{1}{2}}}\cdot\frac{1}{(p^2+1)^{\frac{1}{2}}(q^2+1)^{\frac{1}{2}}}=\frac{\pi}{2}\left(-\frac{\partial}{\partial z}\right)\int_1^\infty\frac{dqe^{-qz}}{(q^2-1)^{\frac{1}{2}}}=\frac{\pi}{2}K_1(z).$$

In (A) and (B) $z=2r$ and $K_\nu(z)$ is a Bessel function of order ν and is related to the well-known Hankel function of the first kind:

$$K_\nu(z)=\tfrac{1}{2}\pi i e^{\nu\pi i/2}H_\nu^{(1)}(z).$$

The asymptotic forms of $K_\nu(z)$ are

for large z: $K_\nu(z)\sim(\pi/2z)^{\frac{1}{2}}e^{-z}$ for all ν,

for small z: $K_0(z)\sim-\log(\frac{1}{2}z)-\gamma$ (γ is Euler constant),

$K_1(z)\sim 1/z$.

All the other integrals which arise may be evaluated by performing suitable differentiations on (A) and (B). Making use of the recurrence relations for the K's, we finally obtain for the interaction potential of any two nuclear particles with spin operators $\boldsymbol{\sigma}_1$ and $\boldsymbol{\sigma}_2$:

$$J_{12}(r)=-3f_1^2\left[\frac{K_0(z)}{z^3}+\frac{2K_1(z)}{z^4}\right]+f_2^2\left[\frac{2K_0(z)}{z^3}+\frac{K_1(z)}{z^2}+\frac{4K_1(z)}{z^4}\right]$$
$$+\frac{f_3^2}{3}\Bigg\{(\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2)\left[\frac{K_0(z)}{z^3}+\frac{2K_1(z)}{z^2}+\frac{2K_1(z)}{z^4}\right]$$
$$+\left[-\frac{3(\boldsymbol{\sigma}_1\cdot\mathbf{z})(\boldsymbol{\sigma}_2\cdot\mathbf{z})}{z^2}+\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2\right]\left[\frac{5K_0(z)}{z^3}+\frac{K_1(z)}{z^2}+\frac{10K_1(z)}{z^4}\right]\Bigg\}$$
$$-\frac{f_4^2}{3}\Bigg\{(\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2)\left[\frac{4K_0(z)}{z^3}-\frac{K_1(z)}{z^2}+\frac{8K_1(z)}{z^4}\right]$$
$$+\left[-\frac{3(\boldsymbol{\sigma}_1\cdot\mathbf{z})(\boldsymbol{\sigma}_2\cdot\mathbf{z})}{z^2}+\boldsymbol{\sigma}_1\cdot\boldsymbol{\sigma}_2\right]\left[\frac{5K_0(z)}{z^3}+\frac{K_1(z)}{z^2}+\frac{K_1(z)}{z^4}\right]\Bigg\}, \quad (7)$$

where $f_i^2=128\pi^3G^2C_i^2$ $(i=1, 2, 3, 4)$, $\mathbf{z}=2\mathbf{r}$.

We now demand of our theory that one type of coupling alone should be capable of explaining all known facts about nuclear forces ("single force hypothesis"; the arguments for this hypothesis are given in Bethe[1]). Then it is clear that the spin-dependence of nuclear forces at once excludes the attractive f_1^2 (scalar) and repulsive f_2^2 (vector) interactions. The f_4^2 (pseudo-vector) interaction can also be eliminated for two reasons: (a) in the singlet state the spin-orbit-spin term vanishes, $\sigma_1\cdot\sigma_2=-3$ and we get for the potential function:

$$J_{12}(r)=f_4^2\left[\frac{4K_0(z)}{z^3}-\frac{K_1(z)}{z^2}+\frac{8K_1(z)}{z^4}\right].$$

For large z, the second term dominates, the potential is attractive and goes as $e^{-z}/z^{2.5}$; for small z the third term is the most important, the potential is repulsive and increases as $1/z^5$. The equilibrium value z_m can easily be found from the minimum condition:

$$K_0(z_m)/K_1(z_m)=(z_m^2+40)/(z_m^3-20z_m);$$

the result is that $z_m=5.8$ which corresponds to $2r_m=5.8\hbar/\mu c$ or a distance $r_m=6.3\times10^{-13}$ cm (for $\mu=175$ electron masses). Such a large lower bound for the region of attractive potential is incompatible with the known short range character of nuclear forces (cf. §1).

(b) The pseudo-vector interaction moreover gives rise to the wrong sign for the quadrupole moment of the deuteron. This can be seen from the fact that the spin-orbit-spin term is repulsive when the vector $\mathbf{z}=2\mathbf{r}$ is parallel to the total spin $\mathbf{S}$. The ground state of the deuteron will consist mostly of a ${}^3D_1$ wave function with an admixture of ${}^3S_1$ and with $\mathbf{z}$ and $\mathbf{S}$ tending to be at right angles. The quadrupole moment will therefore be negative in disagreement with experiment.[10]

There remains only the tensor interaction as a possible theory fulfilling the conditions of the single force hypothesis. It turns out that this interaction is indeed capable of satisfying the criteria for a correct theory enumerated in §1. We write the f_3^2-interaction potential between any two nuclear particles in the form: (cf. Bethe[1] Eqs. (34), (34a), (34b))

$$V=V_1+V_2, \tag{8}$$

where

$$V_1=f^2(\sigma_1\cdot\sigma_2)F(r), \tag{8a}$$

$$V_2=f^2\left[-\frac{3(\sigma_1\cdot\mathbf{r})(\sigma_2\cdot\mathbf{r})}{r^2}+\sigma_1\cdot\sigma_2\right]G(r). \tag{8b}$$

We have used the abbreviations:

$$f^2=f_3^2/24,$$

$$F(r)=\frac{K_0(2r)}{r^3}+\frac{4K_1(2r)}{r^2}+\frac{K_1(2r)}{r^4},$$

$$G(r)=\frac{5K_0(2r)}{r^3}+\frac{2K_1(2r)}{r^2}+\frac{5K_1(2r)}{r^4}.$$

Both $F(r)$ and $G(r)$ are positive monotonically decreasing functions of r—for large r decreasing as $e^{-2r}/r^{2.5}$ while for small r the dominant term becomes $\sim1/r^5$. The exponential decrease of the potential leads to an effective range of the nuclear forces equal to $(\frac{1}{2})\hbar/\mu c\approx1.1\times10^{-13}$ cm.

Comparison of the expression for V as given by (8), (8a), and (8b) with the potential function found by Bethe on the basis of the neutral meson theory shows a striking similarity. Both V_1 and V_2 are identical in the two theories as regards signs, spin factors, etc. except for a different dependence on the separation of the nuclear particles, r. Hence all of Bethe's conclusions about two-body systems, which are independent of the explicit form of $F(r)$ and $G(r)$, hold equally well in the heavy electron pair theory. These conclusions with suitable modification are as follows:

(a) The total angular momentum J, the parity, and the total spin $\mathbf{S}$ are good quantum numbers.

(b) The potential $V_1=-3f^2F(r)$ is attractive at all distances while V_2 vanishes identically for any singlet state of the deuteron or double proton.

(c) For triplet states with $L=J$ the central force V_1 is repulsive; since

$$t_{JL}=\left[-\frac{3(\sigma_1\cdot\mathbf{r})(\sigma_2\cdot\mathbf{r})}{2r^2}+\frac{\sigma_1\cdot\sigma_2}{2}\right]_{JL}=-1$$

for all L, the part V_2 is attractive and independent of L. The total interaction, which is attractive at all distances, is:

$$V_{L=J}=-9f^2\left[\frac{K_0(2r)}{r^3}+\frac{K_1(2r)}{r^4}\right].$$

[10] J. M. B. Kellogg, I. I. Rabi, N. F. Ramsey and J. R. Zacharias, Phys. Rev. **56**, 728 (1939).

(d) The other two triplet states $L=J\pm1$, are always mixed together. For $J=1$ the coupling between the two states $L=2$ and $L=0$ is rather large and will tend to lower appreciably the lower of the two states and to raise the upper. Since $t_{10}=0$ while $t_{12}=1$, the diagonal value of V_2 is more repulsive for the ${}^3D_1$ wave function than for the ${}^3S_1$; hence the ground state of the deuteron if it is a mixture of ${}^3D_1$ and ${}^3S_1$, will be chiefly ${}^3S_1$ rather than ${}^3D_1$ (as in the case of the pseudo-vector interaction). Furthermore it is to be expected that the ${}^3S_1+{}^3D_1$ state will lie below the ${}^3P_1$ state (cf. Bethe[1] §10) and can be made to lie below the ${}^1S$ state by "cutting off" at sufficiently small distances. The ground state of the deuteron will therefore possess a quadrupole moment and in virtue of the attractive character of V_2 for parallel alignment of spin S and vector separation r, the moment will be positive; this is in agreement with experiment.

It is to be noted that in Bethe's theory V_1 has a $1/r$ dependence at small distances so that the $1/r^3$ dependence of the nondiagonal matrix element of V_2 operates to overcome the repulsive action of V_1. In our theory V_1 and V_2 have the same r dependence and the possibility of depressing the triplet state below the singlet arises from the presence of larger numerical coefficients in the nondiagonal matrix element of V_2 than in V_1. Moreover because of the large coupling potential the effect on the energy of the triplet ground state of the deuteron can be considerable without requiring more than a small percentage of D function mixed together with the S function. As far as qualitative considerations go, it would therefore appear that the heavy electron pair theory will give the "almost" additivity of the magnetic moments of neutron and proton.

As has already been implied, the necessity of "cutting off" the potential at small distances exists in the heavy electron pair theory just as it does in Bethe's neutral meson theory. Bethe's arguments for "cutting off" the potential function V (e.g. the interference of the proper fields of the nuclear particles, relativistic corrections, etc.) and his reasons for believing that the saturation requirements for nuclear forces are not in contradiction with the form of V when due account is taken of the "cut-off" (cf. Bethe[1] §6) are equally applicable to the heavy electron pair theory. However these arguments have an admittedly uncertain status at the present time so that we have not attempted any quantitative determination of the "cut-off" distance.

§3. Conclusions

We have seen that the tensor interaction between the nuclear particles and the heavy electron pair field leads to a theory of nuclear forces which is formally identical with the neutral meson theory and is characterized by all the desirable features of the latter theory (cf. §1). In addition, if the single force hypothesis is adopted, the heavy electron pair theory is unique since all the other types of coupling of the nuclear particles to the heavy electron pair field give rise to forces which can be rejected on purely qualitative grounds (cf. §2). If the single force hypothesis is relinquished, many more theories can be constructed by the choice of suitable linear combinations. One can even develop a theory which will lead automatically to repulsion at very small distances and still give the right sign of the quadrupole moment (by a linear combination of the tensor and pseudo-vector interactions). But this procedure involves the introduction of as many (or more) arbitrary constants as there are experimental data to fit.

In the heavy electron pair theory the effective range of nuclear forces is $\frac{1}{2}\hbar/\mu c$ in contrast to a range of $\hbar/\mu c$ in Bethe's neutral meson theory. Insofar as all the results of Breit *et al.* on fitting the experimental data on proton-proton scattering to a meson type of potential function indicate a range of $\frac{1}{2}\hbar/\mu c$, it would seem that this is a definite advantage for the heavy electron pair theory. Of course it is possible that this conclusion will not be borne out by a detailed investigation of the "cut-off" and the strength of the interaction. Whatever be the outcome of such an investigation this paper has established that the existence of unobserved neutral mesons is not indispensable to a theory of nuclear forces.

In conclusion the author should like to thank Professor V. Weisskopf and Dr. A. Nordsieck for helpful discussion.

A Study of the Nuclear Radiations from Antimony and Arsenic

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(Received April 6, 1940)

Beta-gamma and gamma-gamma coincidences were investigated in As^{76}, Sb^{122} and Sb^{124}. Measurements were made on the gamma-ray energies by coincidence absorption of their Compton recoils. Beta-ray endpoints were determined by absorption in aluminum. In As, the measurements indicate three groups of beta-rays all going to excited states of Se^{76}. The endpoint of the highest energy group was found to be 3.24 ± 0.20 Mev. More than one gamma-ray was found to be present. The hardest gamma-ray has an energy of 2.05 ± 0.05 Mev. The effects due to Sb^{122} (2.5 days) and Sb^{124} (60 days) were separated. The beta-ray spectrum of Sb^{124} was found to be simple, consisting of one group going to an excited state of Te^{124}. The endpoint of this group is 1.53 ± 0.05 Mev. More than one gamma-ray was found. The highest energy gamma-ray has an energy of 1.82 ± 0.05 Mev. A soft gamma-ray was also found having an energy less than 69,000 volts. Evidence is offered which suggests the alternative process of K-electron capture with a transition to Sn^{124}. The beta-ray spectrum of the short period Sb^{122} was found to consist of two groups one of which goes to the ground state of Te^{122}. The endpoint of the high energy group was found to be 1.76 ± 0.10 Mev. The endpoint of the lower energy group is estimated at 0.81 Mev. Only one gamma-ray was observed. Its energy is 0.96 Mev. A self-consistent energy level scheme for Sb^{122} is given.

INTRODUCTION

THE study of the radiations emitted from radioactive bodies has usually been carried out by examining the shape of the beta-ray spectrum and measuring the energies and, where possible, the intensities of the various gamma-ray lines emitted. From the results of beta-ray studies alone it is impossible to tell whether the product nucleus is left in the ground state or in some excited state from which it may proceed to the ground state by the emission of gamma-rays. In some cases the beta-ray spectrum appears to be complex and to consist of two or more groups. The maximum energy of each group can be determined with only a fair degree of accuracy. Furthermore, it is generally supposed that the difference in the endpoints of the several groups corresponds to the energy of gamma-rays which are emitted but in many cases correlations have been hard to find. On the other hand, the present technique in the measurement of gamma-rays does not afford very high resolving power so that two gamma-rays lying very close in energy might sometimes be taken for a single gamma-ray. In addition, when methods of gamma-ray measurement are used which are good in the high and medium energy range, low energy gamma-rays may be entirely overlooked. On account of the difficulties involved in the above methods, the number of gamma-rays emitted per disintegration has not, in general, been uniquely determined and the energy level schemes for the various radioactive nuclei are therefore in doubt.

The application of coincidence counting methods to this field has contributed to the solution of many of the problems mentioned above. The principle of the method is to place two counters near the radioactive source and to measure coincidences between (1) the several gamma-rays which may be emitted, and (2) between the gamma-rays and beta-rays of varying energy. If the resolving time of the coincidence circuit is small and the strength of the source is reasonably high, considerable accuracy can be obtained by this method. To date the radiations from As^{76} (26 hours),[1] Mn^{56} (2.5 hours),[2,3] In^{116} (54 minutes),[4] and Na^{24} (14.8 hours)[5] have been investigated. In In and Na it was found that the number of beta-gamma coincidences, calculated per thousand beta-rays, did not change with the energy of the beta-rays, indicating that the beta-ray spectrum of these elements is simple. In Mn, on the other hand, there were found to be proportionately more beta-gamma coincidences associated with elec-

[1] F. Norling, Zeits. f. Physik **111**, 158 (1938).
[2] F. Norling, Naturwiss. **27**, 422 (1939).
[3] L. M. Langer, A. C. G. Mitchell and P. W. McDaniel, Phys. Rev. **56**, 422 (1939).
[4] L. M. Langer, A. C. G. Mitchell and P. W. McDaniel, Phys. Rev. **56**, 380 (1939).
[5] L. M. Langer, A. C. G. Mitchell and P. W. McDaniel, Phys. Rev. **56**, 962 (1939).

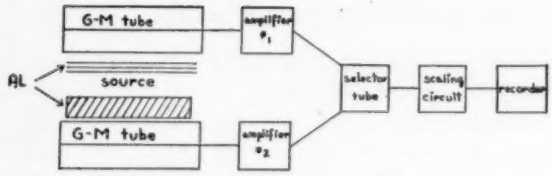

FIG. 1. Schematic arrangement for counting beta-gamma and gamma-gamma coincidences.

trons of low energy, showing that there are two groups of beta-rays in Mn and that the lower energy group is accompanied by a gamma-ray which is not found with the high energy electrons. In all three cases, beta-gamma coincidences were found to persist all the way to the endpoint and in addition gamma-gamma coincidences were found. These facts show that, in these elements, there is more than one gamma-ray per disintegration and that the process of beta-ray emission leaves the product nucleus in an excited state. Norling's results on As were obtained with such a weak source that it is difficult to draw any definite conclusions from this work. The experiments are repeated in the present article.

In the present experiments, measurements of the coincidence type have been made on As, the two periods of Sb (2.5 days) and (60 days), Eu, and Dy. In addition the energies of certain gamma-rays in As and Sb have been determined as well as the beta-ray endpoints of the various substances.

1. APPARATUS

The coincidence counting apparatus was used in two different modifications for the purposes of (1) measuring coincidences between beta- and gamma-rays and those between two or more gamma-rays, respectively; and (2) of determining the energy of gamma-rays by measuring the range of Compton electrons produced by them in aluminum.

The apparatus for measuring coincidences between beta- and gamma-rays is shown in Fig. 1. Two Geiger-Müller tube counters, 2 cm in diameter and 6 cm long, were placed with their centers 4.5 cm apart in a horizontal plane. The source of radioactive material was placed midway between the two counters. Between the source and one of the counters, usually designated as the gamma-ray counter, there was placed a block of aluminum of dimensions large enough to shield the counter from the radiations emitted by the source and of thickness great enough to stop all beta-rays coming from the source. In those instances in which coincidences between gamma-rays were to be measured, a similar block of aluminum was located between the source and the other counter. In those cases in which coincidences between beta-rays and gamma-rays were to be measured, there was placed between the source and the second counter, designated as the beta-ray counter, thin sheets of aluminum of thickness sufficient to stop beta-rays of energy less than a certain specified amount. The thickness of these sheets was varied from experiment to experiment so that the number of coincidences between beta- and gamma-rays could be determined as a function of the energy of the beta-rays. The counter walls had a stopping power equivalent to 0.05 g/cm² of aluminum. The counters themselves were filled with a mixture of 8 cm of argon and 1 cm of ethyl alcohol. The pulses from the counter were fed into an improved resistance-capacity coupled coincidence amplifier similar to one previously described[6] and thence to the scaling circuit and recorder. The apparatus was capable of counting with negligible losses at rates up to 30,000 per minute. The time constants of the apparatus were reduced in such a manner as to give a high resolving time for coincidences but at the same time to maintain the desirable feature of having all the pulses reaching the mixer tube of the same size. The resolving time for coincidences was determined by recording the chance coincidences when an independent source of beta-rays was placed over each counter.[3]

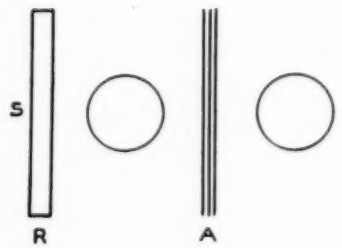

FIG. 2. Schematic arrangement for measuring gamma-ray energies by absorption in A of Compton recoils produced in R by a source S.

[6] L. M. Langer and M. D. Whitaker, Phys. Rev. **51**, 713 (1937).

Throughout these experiments the resolving time, τ, was 0.54×10^{-7} min.

Before and after each coincidence run single counts were taken in each counter so that the chance rate could be determined. Before any coincidence measurements between beta- and gamma-rays were made, the gamma-gamma coincidence rate was determined so that this could be subtracted, together with the chance rate, in determining the true beta-gamma coincidence rate.

The arrangement for measuring the energy of the gamma-rays is shown in Fig. 2. Gamma-rays from the radioactive source, S, eject Compton electrons from the aluminum radiator, R. These electrons then go through both counters, being recorded as coincidences. Sheets of aluminum are then placed at the point A between the counters to absorb the electrons. The number of coincidences per minute is then measured as a function of the thickness of the absorber A.

The energy of the *maximum energy* gamma-ray may be determined by observing that thickness of aluminum which completely stops all coincidences above the background rate. The energy of these electrons E may then be calculated from their range in aluminum taking into account, of course, that they have had to traverse the thickness of three counter walls or 0.15 g/cm² of aluminum in addition to the absorber thickness. The energy of the gamma-ray $h\nu$ may best be calculated with the help of the curve published by Curran, Dee and Petrzilka[7] which was obtained by measuring the range of Compton electrons ejected by gamma-rays of known energy.

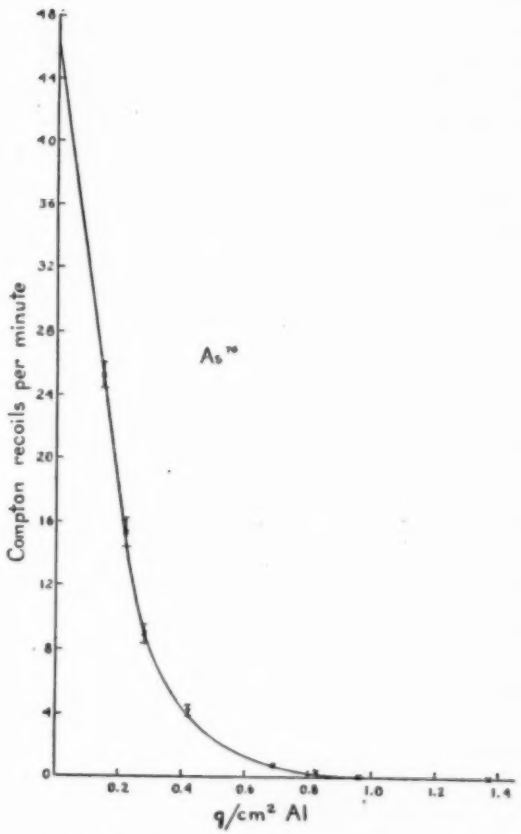

FIG. 4. Coincidence absorption of Compton recoils produced by gamma-rays of As.

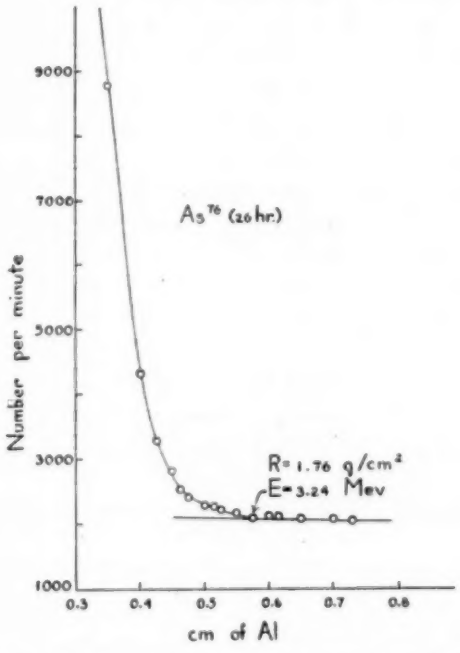

FIG. 3. Absorption in Al of beta-rays from As⁷⁶.

The *average energy* of the gamma-rays, if there is more than one, can be obtained by measuring the thickness of aluminum necessary to diminish the coincidence rate to one-half its value with no absorber present. If $D_{\frac{1}{2}}$ is the thickness in g/cm² necessary to cut the rate to one-half then[8,9]

$$D_{\frac{1}{2}}=C_{\frac{1}{2}}\frac{(h\nu/mc^2)^2}{(h\nu/mc^2)+1}. \qquad (1)$$

[7] S. C. Curran, P. I. Dee and V. Petrzilka, Proc. Roy. Soc. **169**, 269 (1938).
[8] F. Rasetti, Zeits. f. Physik **97**, 64 (1935).
[9] A. C. G. Mitchell and L. M. Langer, Phys. Rev. **52**, 137 (1937).

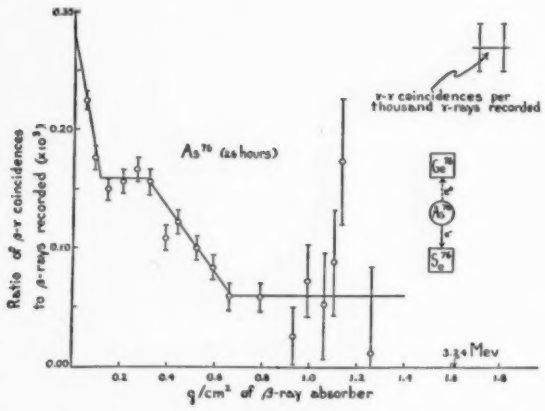

FIG. 5. Beta-gamma coincidences in As^{76} as a function of beta-ray energy.

The value for $C_{\frac{1}{2}}$ is determined by calibration with a known gamma-ray and has been shown to be[9] 0.0650 g/cm². A similar result holds for the quarter-value thickness, with $C_{1/4}$ taken as 0.110 g/cm². In this work both methods were used and the results compared with each other in order to see how many gamma-rays might be present.

2. MEASUREMENTS ON As^{76}

A strongly activated sample of As^{76}, produced by bombarding arsenic with slow neutrons from the Berkeley cyclotron, was used throughout this work. During the course of the experiments, counting rates in the beta-ray counter and the gamma-ray counter were made separately at frequent intervals so that enough data were obtained to give an accurate value of the period. From these data, taken over a period of eight half-lives, a value of 26.3 ± 0.3 hours was obtained for the half-life, which is in excellent agreement with that obtained by other observers.

The endpoint of the beta-ray spectrum was determined by measuring the number of beta-ray counts as a function of the thickness of aluminum absorbers placed between the source and one of the counters. The results are shown in Fig. 3 from which the maximum range, including the thickness of the counter wall, was found to be 1.76 g/cm² of aluminum. From Feather's rule, this corresponds to a beta-ray endpoint of 3.24 $\pm$0.20 Mev. This is to be compared with an extrapolated K.U. endpoint obtained by Brown and Mitchell[10] of 3.4 Mev, a similarly determined value of 3.16 ± 0.10 by Harteck, Knauer and Schaeffer,[11] and an observed endpoint of 2.71 $\pm$0.14 obtained by Weil and Barkas.[12]

The energy of the gamma-radiation was next examined by the method of absorbing the Compton electrons ejected from aluminum as described above. A curve showing the coincidence rate as a function of total absorber thickness is shown in Fig. 4. The maximum range of the Compton electrons is 0.95 ± 0.05 g/cm². From the data of Curran, Dee and Petrzilka,[7] the value of the gamma-ray energy is found to be 2.05 ± 0.05 Mev. This, of course, represents the energy of the hardest gamma-ray. The value of the gamma-ray energy determined from the half-value thickness is 1.67 Mev, and from the quarter-value thickness is 1.62 Mev. Since the values obtained from the half- and quarter-value thicknesses give only the average energy of all the gamma-rays, it would appear that there are other gamma-rays of energy less than 2.05 Mev present. This supposition is borne out by the fact that we have found gamma-gamma coincidences in arsenic. The gamma-ray of energy 2.05 Mev found in this work agrees quite well with that of energy 2.16 Mev found by Harteck, Knauer and Schaeffer in their cloud-chamber experiments. The average energy of the gamma-rays obtained from the half-value thickness indicates that the gamma-ray of energy 1.5 Mev found by these authors is probably also present. The 3.15-Mev line mentioned by them was not found in the present work.

The sample was now placed in the coincidence counting arrangement and investigated for coincidences between gamma-rays. A thickness of aluminum (0.73 cm) sufficient to stop all beta-particles was placed on each side of the source. In a series of three experiments in which from 600 to 1400 coincidences were counted, the number of gamma-gamma coincidences per thousand gamma-ray counts was found to be 0.27 ± 0.02.

Experiments were next performed in which the

[10] M. V. Brown and A. C. G. Mitchell, Phys. Rev. **50**, 593 (1936).
[11] P. Harteck, F. Knauer and W. Schaeffer, Zeits. f. Physik **109**, 153 (1938).
[12] G. L. Weil and W. H. Barkas, Phys. Rev. **56**, 485 (1939).

number of beta-gamma coincidences were measured as a function of the thickness of aluminum between the source and the beta-ray counter. Before and after each run the number of singles in the beta-counter and that in the gamma-counter were measured. In addition the number of gamma-ray counts in the beta-counter was also determined. From these data the number of beta-ray counts in the beta-ray counter could be determined at the mean time of the run. The number of true beta-gamma coincidences per thousand beta-ray counts was then calculated. The results are shown in Fig. 5, in which the number of beta-gamma coincidences per thousand beta-rays recorded is plotted as a function of the thickness of absorbing material traversed.

It was difficult to obtain accurate results in the neighborhood of the endpoint since the number of beta-rays recorded was small and had to be determined in the presence of a large background of gamma-ray counts. The results indicate, however, that there are beta-gamma coincidences out to the endpoint of the beta-ray spectrum. As one proceeds from the lower energy end of the spectrum it is apparent that the number of beta-gamma coincidences decreases as electrons of low energy are cut out. This occurs in two well-defined groups, one of which is associated with electrons of energy less than 0.5 Mev (0.11 g/cm²), and the other with electrons of energy less than 1.5 Mev (0.66 g/cm²). The beta-ray spectrum of As^{76} should therefore consist of three beta-ray groups—one with an endpoint of about 0.5 Mev, another at 1.5 Mev and the third with an endpoint of 3.24 Mev. In addition the highest energy beta-ray does not lead to the ground state of Se^{76} since beta-gamma coincidences were found with this group as close to the endpoint as reliable results could be obtained.

The authors do not feel that a quantitative energy level scheme for Se^{76} can be drawn up from these data since the curve shown in Fig. 5 is an integral curve and it is impossible to determine the inner endpoints with any accuracy. Qualitatively, however, one can see that the nucleus of Se^{76} must consist of three levels above the ground level. The three upper levels should be approximately 0.5, 1.5, and 3.2 Mev below the ground level of As^{76}. The relation of the lowest of these levels and the ground level of Se^{76} is not at present known. This level scheme is in qualitative agreement with the one published by Schaeffer and Harteck[13] except that they assumed that the 3.2-Mev beta-ray led directly to the ground state of Se^{76}. In addition, this scheme would provide for several gamma-rays, probably four. One of these gamma-rays, of energy 2.05 Mev, has been definitely established in this work, and evidence for others of lower energy has been obtained.

It should be noted that As^{76} has such a position in the periodic table that it may decay to Ge^{76} by positron emission or K-electron capture, or to Se^{76} by electron emission. Evidence for the emission of positrons was found by Harteck, Knauer and Schaeffer.[11,13] The number of positrons found, however, was only about one percent of the number of electron tracks. In the present experiments we have made no attempt to separate the positrons from the rest of the electrons, but we feel that this small number of positrons would not affect our results appreciably.

Finally, it should be pointed out that the ratio of the number of gamma-gamma coincidences per recorded gamma-ray to the number of beta-gamma coincidences per recorded beta-ray is much higher in these experiments than we have found in the elements that we have hitherto investigated. This indicates that there are a considerable number of gamma-rays per disintegration, some of which are presumably not accompanied by the emission of a beta-ray. The ratio is approximately unity when electrons of all energies are considered. The value of 0.27×10^{-3} gamma-gamma coincidences per gamma-ray is to be compared with the extrapolated value of 0.28×10^{-3} beta-gamma coincidences per beta-ray as shown in Fig. 5. It can be shown that (see Section 5 on theory), in the region in which the above-mentioned ratio is near unity, K (the number of gamma-rays per disintegration) cannot be determined with any accuracy, since the value of K is extremely sensitive to small variations in the value of this ratio.

[13] W. Schaeffer and P. Harteck, Zeits. f. Physik **113**, 287 (1939).

3. Measurements on Antimony

Antimony has two stable isotopes, Sb^{121} and Sb^{123}, from which two radioactive isotopes can be formed by the capture of slow neutrons. Both radioactive isotopes are electron emitters and their periods have been determined as 2.8 days for Sb^{122} and 60 days for Sb^{124} by Livingood and Seaborg.[14] The endpoint of the beta-ray spectrum of Sb^{122} has been determined with the help of a cloud chamber by Amaki and Sugimoto[15] who obtained a value of 1.64 Mev. In addition gamma-rays have been reported from the two isotopes by Livingood and Seaborg.

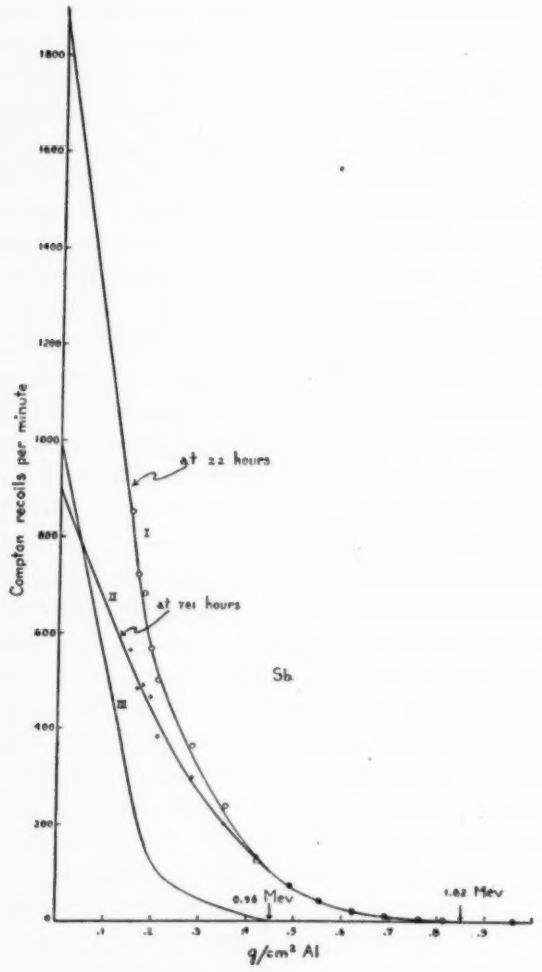

Fig. 6. Coincidence absorption of Compton recoils produced by gamma-rays of Sb^{122} and Sb^{124}.

[14] J. T. Livingood and G. T. Seaborg, Phys. Rev. **55**, 414 (1939); Rev. Mod. Phys. **12**, 30 (1940).
[15] T. Amaki and A. Sugimoto, Inst. Phys. and Chem. Research, Tokyo, Sci. Paper No. **853**, 1650 (1938).

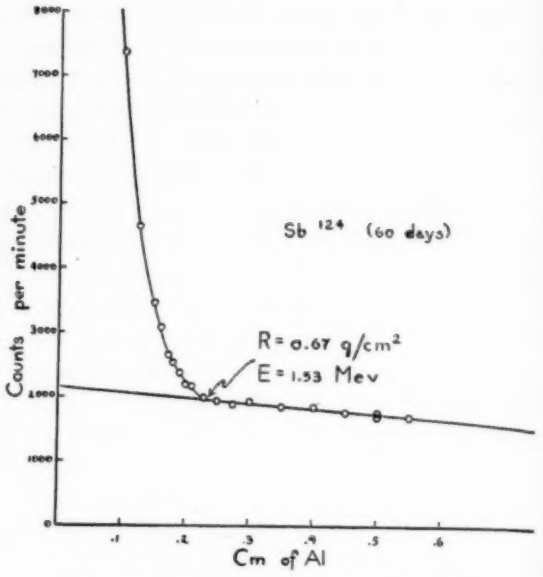

Fig. 7. Absorption in Al of beta-rays of Sb^{124}.

Through the courtesy of the Radiation Laboratory in Berkeley, a sample of antimony was strongly irradiated with slow neutrons and shipped back to us for study. Upon investigation it turned out that both periods were strongly activated. We originally hoped that coincidence measurements taken at various times throughout the life of the sample would enable us to determine the effects due to both periods, but this turned out to be impossible since the points at low absorber thickness gave such a large count that they could not be determined until most of the activity due to the short period had died out. After measurements on the long period had been completed we obtained a sample from the University of Michigan, which had been irradiated for only two hours, in which the ratio of the beta-activity of the short period to that of the long period was about 30 to 1.

No attempt was made in this work to determine the half-life of the long period with any accuracy. Our points agree reasonably well with a half-life of 60 days as determined by others. Measurements on the short period, taken with greater care, give a value of 63 hours for its half-life.

Measurements of the energy of the gamma-rays from the two periods were made in the manner described above. Two series of measure-

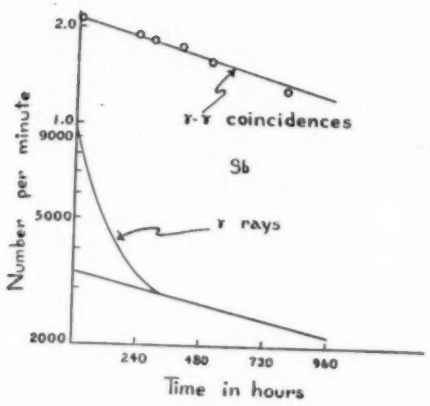

FIG. 8. Decay curve for the gamma-rays of Sb showing gamma-gamma coincidences with long period only.

ments were made with the strongly activated sample: one set taken at 22 hours after receiving the sample gave a composite curve for both periods, while the other, taken at 781 hours after the start of the work, gave results which are attributable to the long period only. These two curves, adjusted for decay of the source, are shown as curves I and II of Fig. 6. The difference curve (curve III), represents the measurements on the gamma-rays of the short period.

The maximum range for the Compton electrons from the long period is 0.852 g/cm², corresponding to a gamma-ray energy[7] of 1.82 ±0.05 Mev. The energy of the gamma-ray determined from the half- and quarter-value thicknesses with the help of Eq. (1) is 1.85 and 1.89 Mev, respectively. One would therefore conclude that, if there are more than one gamma-ray of energy greater than 1 Mev, they are so closely spaced about 1.8 Mev as not to be resolved by this method. It will be shown later that there is another gamma-ray of much lower energy present.

The maximum range of the Compton electrons from the gamma-ray of the short period is 0.45 g/cm², corresponding to a gamma-ray energy of 0.96 Mev. The half- and quarter-value thicknesses give 1.15 Mev and 1.10 Mev, respectively. Here again if there are more than one gamma-ray they must be closely spaced about 1.0 Mev.

a. Coincidence measurements on the long period

As a preliminary experiment the endpoint of the beta-ray spectrum of the long period was determined by measuring the range of the electrons in aluminum with a single counter. The results are shown in Fig. 7, in which the number of counts per minute is plotted as a function of the thickness of the aluminum absorber. The maximum range corresponds to a beta-ray energy of 1.53±0.05 Mev. It should also be noted that the gamma-ray background decreases with increasing thickness of aluminum from 0.2 cm to 0.6 cm, indicating the presence of a low energy gamma-ray readily absorbed in aluminum.

With 0.6 cm of aluminum between the source and each counter, measurements of gamma-gamma coincidences were made. These observations were carried out on many occasions from the time that the source was received until it was 800 hours old. At the time of each measurement of the coincidence rate the gamma-ray counting rate in a single counter was also measured. The results of these experiments are shown in Fig. 8 in which both the gamma-gamma counting rate and the intensity of the gamma-rays, as recorded in a single counter, are shown as a function of the time. It will be seen at once that, whereas the intensity as measured with the single counter, decays in a manner which is a composite of both periods, the coincidence counting rate decays with the half-life of the long period. This is true even in the early part of the run when the intensity from the short period is approximately three times that from the long period. This indicates at once that there are no gamma-gamma coincidences connected with the short period.

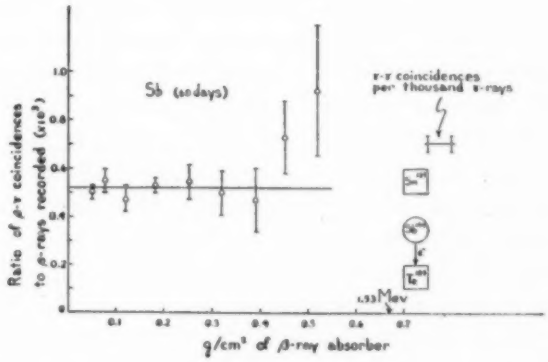

FIG. 9. Beta-gamma coincidences in Sb^{121} as a function of beta-ray energy.

After the short period activity had died out of the source beta-gamma coincidences were measured as a function of the thickness of aluminum between the source and the beta-ray counter, keeping 0.60 cm of aluminum between the source and the gamma-ray counter. The results are shown in Fig. 9, in which the number of beta-gamma coincidences per beta-ray recorded is plotted as a function of the thickness of aluminum absorber. On the same diagram is plotted the number of gamma-gamma coincidences per gamma-ray recorded when 0.60 cm of aluminum is placed between the source and each counter. All results are, of course, corrected for decay and chance counting rate in the usual manner.

It will be noticed that the number of beta-gamma coincidences per recorded beta-ray is independent of the energy of the beta-rays over the entire range which we were able to measure. The value of this ratio is 0.52×10^{-3} coincidence per recorded beta-ray. These results indicate that the shape of the beta-ray spectrum of Sb^{124} is simple and that the resulting Te^{124} nucleus is left in an excited state.

The number of gamma-gamma coincidences per recorded gamma-ray is 0.70×10^{-3} and is greater than the value 0.52×10^{-3} beta-gamma coincidence per recorded beta-ray by a factor of 1.35. As will be shown in Section 5, this result cannot be explained on the hypothesis that the parent nucleus emits a beta-ray which is followed by several gamma-rays. Since Sb^{124} may be transformed into Te^{124} by electron emission or into Sn^{124} by K-electron capture, it appears likely that the second process is also taking place. If this were the case there would be gamma-rays emitted in the rearrangement of the Sn nucleus which are not associated with beta-emission, thus accounting for the high value of the gamma-gamma coincidence rate compared to that due to beta-gamma coincidences.

It appeared of interest to obtain more information on the gamma-ray of low energy which was observed at the tail of the beta-ray absorption curves. A new counter holder was therefore made in which the two counters were 6.5 cm apart between centers. With this apparatus it was possible to go to greater absorber thickness so

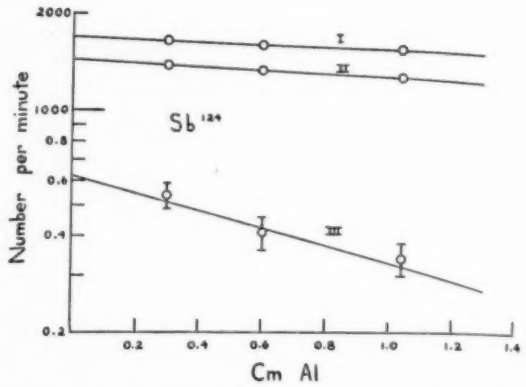

FIG. 10. Absorption in Al of gamma-rays of Sb^{124}. Curves I and II show absorption of singles. Curve III shows absorption of gamma-gamma coincidences.

that more points on the gamma-ray absorption curve could be obtained. In addition, with this symmetrical arrangement, the absorption of the gamma-gamma coincidences in various materials could be observed. The object of these experiments was to obtain a measure of the energy of the low energy gamma-ray and also to ascertain whether the low energy gamma-ray was involved in the gamma-gamma coincidences which were previously observed.

One can readily see that if there are two gamma-rays present the absorption curve of the singles will be given in the usual manner by

$$N_\gamma = N[S_{\gamma_1}{}^0 e^{-\mu_1 x} + S_{\gamma_2}{}^0 e^{-\mu_2 x}]$$

and the absorption coefficient of the low energy gamma-ray will have to be measured in the presence of a background due to the high energy gamma-ray. On the other hand, if the low energy gamma-ray is coincidental with another high energy gamma-ray, it is clear that if the soft gamma-ray is recorded in one counter the high energy one will be recorded in the other and vice versa, so that the intensity of the coincidences should be governed by the absorption coefficient of the softest gamma-ray. The absorption curve is then expressible as

$$N_{\gamma\gamma} = 2NS_{\gamma_1}{}^0 S_{\gamma_2}{}^0 e^{-(\mu_1+\mu_2)x},$$

where $S_{\gamma_1}{}^0$ and $S_{\gamma_2}{}^0$ are the sensitivities of the counters for the gamma-rays 1 and 2, and where μ_1 and μ_2 are the absorption coefficients of the two gamma-rays.

The experiments were carried out with both lead and aluminum as absorbers, the coincidence curve being obtained by placing an equal amount of absorber on each side of the source. The results are shown in Fig. 10. Curves I and II show the intensity of the singles as a function of thickness of an aluminum absorber, while curve III is a similar curve for coincidences. It is apparent at once that the low energy gamma-ray is involved in the coincidences and that the absorption coefficient of the soft gamma-ray is dominating the coincidence curve. The mass absorption coefficient obtained for the aluminum curve is 0.232 cm^2/g which corresponds to an energy of about 69,000 volts. Since the geometry of this arrangement was not particularly suitable for absorption coefficient work, the source subtending too large a solid angle at the counters, the value obtained for the mass absorption coefficient is a lower limit and the energy of the gamma-ray therefore an upper limit. Experiments in which lead was used as an absorber gave results in qualitative agreement with those mentioned above.

Beta-gamma coincidences were also measured in such a manner that there was no absorber between the source and the beta-ray counter, and varying thicknesses of aluminum were placed between the source and the gamma-ray counter. In this way the number of beta-gamma coincidences was obtained as a function of the absorption of the gamma-rays. In this case the absorption was slight and was definitely not so large as that for the gamma-gamma coincidences. It therefore appears that the beta-rays are not coincidental with a low energy gamma-ray alone. One can conclude from these experiments that the beta-rays are coincidental with a high energy gamma-ray alone or with a combination of a high energy and low energy gamma-ray.

b. Coincidence measurements on the short period, Sb^{122}

In order to make measurements on the 63-hour period, Sb^{122}, the source which had been activated for two hours, was used. Throughout the time necessary to make the measurements the ratio of the activity due to Sb^{122} to that of Sb^{124} was about 30 to 1.

The absorption of the beta-rays was measured in a single counter and a range of 0.794 g/cm^2 of aluminum was obtained. This value corresponds to an endpoint energy of 1.76 ± 0.10 Mev, which agrees, within the limit of error, with that found by Amaki and Sugimoto[15] and is somewhat greater than the value of 1.53 Mev found by us for Sb^{124}.

The number of gamma-gamma coincidences was found to be quite small and could be accounted for exactly from the small amount of the long period existing in the source. This result is in agreement with the fact that the gamma-gamma coincidences obtained from the other source were found to decay with a period of 60 days. Since there are no gamma-gamma coincidences in Sb^{122} and since we have shown that there is a gamma-ray present, it follows that there is only one gamma-ray per disintegration.

The number of beta-gamma coincidences per recorded beta-ray is plotted in Fig. 11 as a function of the thickness of aluminum absorber between source and beta-ray counter. It will be seen that there are no beta-gamma coincidences recorded when all electrons of range less than 0.28 g/cm^2 are cut out of the spectrum, but for the lower energy electrons there is a rise in the number of beta-gamma coincidences per recorded beta-ray. This indicates that the beta-ray spectrum of Sb^{122} consists of two groups, the most energetic of which leads to the ground state of Te^{122}, and the other to an excited state from which a gamma-ray may be emitted. From Fig. 11 the endpoint of the lower energy group may be determined with fair accuracy as 0.81 Mev.

The data obtained so far enable one to construct a reasonable energy level scheme for the process $Sb^{122} - Te^{122}$. This scheme is also shown in Fig. 11. The highest energy beta-ray group has an endpoint of 1.76 Mev which leads directly to the ground state of Te^{122} since no gamma-gamma coincidences are found. In addition there is a lower energy group of beta-rays having an endpoint of 0.81 Mev and one gamma-ray whose energy we found to be 0.96 Mev. The sum of the energies of the low energy beta-ray and the gamma-ray is equal to that of the high energy beta-ray within the limits of experimental error.

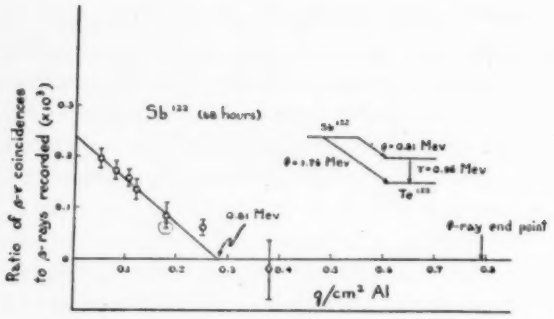

FIG. 11. Beta-gamma coincidences in Sb^{122} as a function of beta-ray energy.

4. EXPERIMENTS ON Eu AND Dy

A sample of dysprosium oxide was irradiated with neutrons from a 200 mg radium-beryllium neutron source until the 2.5-hour period was strongly activated. Measurements showed that the gamma-ray activity, if any, was extremely weak, there being only 35 counts per minute recorded in the gamma-ray counter when there were 13,000 per minute in the beta-ray counter.

A sample of europium oxide was kindly sent us by Professor Cork. This sample was the one that had previously been used for the work on the very long period of Eu and still showed considerable activity. The sample showed some evidence of beta-gamma coincidences but its activity was not great enough to allow us to make precise measurements. The sample was then activated with the radium-beryllium neutron source with the hope of obtaining data on the 9.2-hour period. Here again the number of gamma-rays due to this period was small compared to the number of beta-rays so that coincidence work was impossible.

5. DISCUSSION OF THE THEORY

In order to find the number of gamma-rays per disintegration from coincidence experiments with two counters one may make the following considerations. Let there be N_β disintegrations per second and let K be the average number of gamma-rays per disintegration. Let S_β and S_γ be the sensitivity of the beta-ray counter and gamma-ray counter, respectively, the solid angle subtended by the counter being included. In addition suppose that S_γ is essentially constant over the region of gamma-ray energies investigated. The number of beta-gamma coincidences per second will be

$$N_{\beta\gamma}=N_\beta S_\beta \cdot KS_\gamma$$

and the number of beta-gamma coincidences per recorded beta-ray will be

$$N_{\beta\gamma}/N_\beta S_\beta = KS_\gamma.$$

If gamma-gamma coincidences are to be observed, then if one gamma-ray is recorded in one counter any one of $K-1$ will be recorded in the other. Since there are K gamma-rays per disintegration the number of gamma-gamma coincidences per second will be

$$N_{\gamma\gamma}=N_\beta KS_\gamma \cdot (K-1)S_\gamma.$$

The number of gamma-gamma coincidences per recorded gamma-ray will therefore be

$$N_{\gamma\gamma}/N_\gamma S_\gamma = (K-1)S_\gamma.$$

In the case of a simple beta-ray spectrum, in which the number of beta-gamma coincidences per recorded beta-ray is independent of the energy of the beta-rays, the ratio of the number of gamma-gamma coincidences per recorded gamma-ray to the number of beta-gamma coincidences per recorded beta-ray is given by

$$R=\frac{N_{\gamma\gamma}}{N_\gamma S_\gamma}\Big/\frac{N_{\beta\gamma}}{N_\beta S_\beta}=\frac{K-1}{K},$$

so that K may be calculated from this ratio.

The maximum value of the ratio R cannot be greater than unity and still comply with the suppositions on which the theory is based. For values of R greater than unity it is clear that other effects are coming in which were not taken into account in deriving the formula. This can only be the emission of gamma-rays not connected with recorded beta-rays. In the case of Sb^{124} for which the value of R is 1.35, it would seem that the only reasonable way to account for this value would be to assume that in a certain fraction of the transitions Sb^{124} is being transformed into Sn^{124} by K-electron capture, and in the resulting rearrangement of the Sn nucleus gamma-rays are given out.

The authors wish to express their appreciation

to Professor E. O. Lawrence and Dr. E. Segrè of the Radiation Laboratory of the University of California and also to Professor J. M. Cork of the University of Michigan for furnishing them with the strong sources used in these experiments. They are also indebted to Professor E. J. Konopinski for many helpful discussions during the progress of the work. They also wish to express their thanks to the Penrose Fund of the American Philosophical Society for a grant.

JUNE 15, 1940 PHYSICAL REVIEW VOLUME 57

Protons from the Transmutation of Boron by Deuterons

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(Received April 8, 1940)

The technique of studying proton groups produced by bombarding elements with deuterons from a cyclotron is described. Applied to the bombardment of boron by 3.1-Mev deuterons, the method shows the presence of four groups, three of which agree with those found by Cockcroft and Lewis while the fourth, a very prominent group of Q value +2.39 Mev is new and is ascribed to a third excited state of B^{11}.

INTRODUCTION

THE earliest experiments performed with the cyclotron were studies of charged particles emitted in nuclear reactions. However, since the discovery of artificial radioactivity for the production of which the cyclotron is so powerful an instrument, the emphasis in cyclotron work has shifted and little work with charged particles has been carried out. The cyclotron at the Sloane Physics Laboratory was designed to employ the beam mainly in direct studies of transmutations, rather than in production of therapeutic samples of radioactive materials. For this purpose a moderate sized cyclotron giving relatively small ion currents is quite satisfactory. Such a cyclotron has been developed at low cost of construction and put into operation. As a first experiment boron was bombarded, since the excellent work of Cockcroft and Lewis[1] affords a means by which the interpretation of results can be checked. The agreement found is good and further additional information was added by the work.

EXPERIMENTAL EQUIPMENT

The cyclotron magnet has 28″ pole pieces, designed to fit the 27½″ acceleration chamber formerly in use at Berkeley.[2] The authors are highly appreciative of the opportunity to use this chamber which has proved quite satisfactory and, once the exigencies of large scale waxing were met, has given very little trouble. The source of R.F. power is a master oscillator-amplifier with an output stage consisting of four air-cooled RCA-833 tubes in parallel push-pull. We have been able to obtain one microampere of 3.1-Mev deuterons with a total input power of 3.5 kw. Of this a little over 1 kw appears in the cooling water flowing through the dees. For most of this work one microampere is more than enough, as it causes heating in volatile targets which are harmless with less intense bombardment.

The beam is deflected into an external bombardment chamber separated from the acceleration chamber proper by a brass wall and strikes a target of the element to be studied which is placed at 45° to the beam. A hole of 1 cm diameter, 8 cm from the target, covered with an aluminum foil, permits the protons ejected from the target at 90° to the incident beam to escape. The area of target struck by the beam varies somewhat with the conditions of operation of the cyclotron (for example, upon shims, and R.F.

[1] J. D. Cockcroft and W. B. Lewis, Proc. Roy. Soc. **154**, 261 (1936).

[2] E. O. Lawrence and D. Cooksey, Phys. Rev. **50**, 1131 (1936).

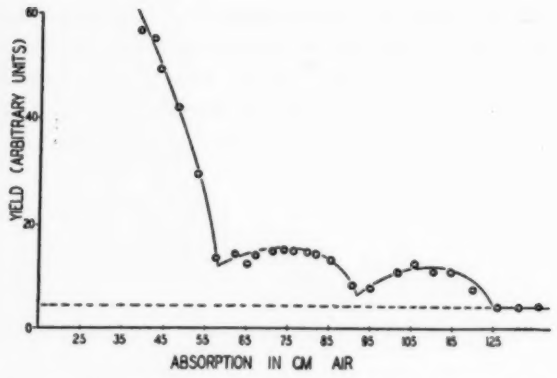

FIG. 1. Yield of protons plotted against absorption for the region covered by the work of Cockcroft and Lewis. Three groups are observed in good agreement with their values. The observed relative yields are different because of the higher excitation energy in these experiments.

voltage) but is remarkably small, and in general is less than one square centimeter. The total angular spread in these experiments is therefore about 8° or 4° each side of the mean value. For experiments with deuterons in which the variation of range with angle is not great this spread is quite small enough to ensure accuracy in measurement of energy values.

The protons and alpha-particles emitted by the target are detected by means of a proportional counter[3] as described for work with natural sources. A linear amplifier and ionization chamber can be used also, but experience has shown that for fast proton detection the proportional counter is more reliable and less affected by electrical disturbances. The absorption curves were plotted by interposing aluminum foils. These were placed in two sliders, each carrying six foils, thus permitting 36 possible combinations to be selected rapidly. The foils were measured and weighed and it was assumed that 1.52 milligrams per square centimeter is equivalent to one centimeter of air. Corrections were applied as described by Livingston and Bethe and the Cornell range-energy relations were used in reducing the data.

The maximum energy of the deuterons was measured in three ways: first, by letting the beam escape through a foil into the air and measuring its range, second, by passing the beam into a chamber into which absorbing gas could be admitted at various pressures and so determining its range, and third, by observing the energy of protons of known origin produced in a thin target. All three methods agreed, giving a value of 3.1 Mev. The beam has a clearly marked maximum and is reasonably homogeneous. Definite ranges are produced when elements are bombarded. It is considerably more homogeneous than most polonium sources. (The spread in range is estimated as ±0.7 cm or ±0.3 Mev.)

In experiments with deuterons, target contamination must be guarded against. This is not so serious when more energetic deuterons are used as it is with, say, 0.5-Mev deuterons. For deuterons of high energy the cross section of the element studied is generally almost as large as that of a light element contaminant, so that any spurious yield is proportional to the amount of impurity and not greatly favored by a larger cross section. Hence if reasonably pure targets are used the danger of erroneous results due to contaminants is not great. The commonest and in fact almost universal contaminants are carbon, from the pump oil, and oxygen from general handling. These elements do not interfere in experiments employing a thick layer of boron which gives a large yield itself. In work with elements such as copper or vanadium, however, they offer a serious difficulty, so serious that only groups having ranges beyond 34 cm (carbon) are considered as being due to the target proper unless there is good reason to discount the presence of carbon impurity. We found that targets having a smooth surface do not readily acquire carbon contamination, but that a copper target rapidly becomes blackened and would do very well for a thin carbon target.

EXPERIMENTAL RESULTS

Boron layers were prepared by painting an alcohol suspension of the element on to a gold backing. The layers were not uniform but reasonably thin so that the greatest thickness of a thin layer was less than one centimeter air equivalent. Many absorption curves were plotted for both thin and thick layers: of these, two are shown in Figs. 1 and 2. In Fig. 1 the groups occurring between absorptions of 35 and 135 cm

[3] D. G. Brubaker and E. Pollard, Rev. Sci. Inst. 8, 254 (1937).

are shown. It will be seen that three groups are present, having ranges of 125, 92, and 58 cm, (uncorrected for variation of range in Al with velocity) with a possible error of ±2 cm. The ordinates are the yield of protons per centimeter deflection of the beam meter. The amount of beam was estimated by visual observation of the meter, a procedure rendered possible by the steadiness of the beam which is constant on most occasions to better than ten percent so that the visual average is correct to five percent at worst. A photographic recorder was used to test the accuracy of the observer and showed that the steadiness of the beam warranted the use of the visual average. In later experiments a current integrator has been used and a comparison between "visual" runs and those taken with the integrator shows excellent agreement. The particles were counted with a "scale-of-ten" circuit as described by Kerst[4] and over a thousand particles were counted at each point; for high yields a great many more. The points were not taken consecutively but more or less at random and they have not been adjusted in any way so that the agreement between nearby points shows the essential steadiness of the conditions of work. The energy changes for these three groups are calculated to be +9.22, +7.30 and +5.00 Mev, each with a probable error of 0.2 Mev. The values found by Cockcroft and Lewis are 9.14, 7.00 and 4.71 Mev, which agree reasonably well with our figures. We have not made any great attempt to push the accuracy of range measurement to the limit in this case since the values of Cockcroft and Lewis are satisfactory. The general trend of our values to be slightly lower than theirs may be significant. Our value for the mass of B^{11} if we assume 10.01579 for B^{10} as suggested by Allison,[5] is 11.0126±0.0002, which is in agreement with the value he proposes in the same paper.

In Fig. 2 is shown the absorption curve for protons between 16 cm and 35 cm which would have ranges masked by heavy alpha-particle groups in the experiments of Cockcroft and Lewis but which become apparent at our higher bombarding energies. A very definite large yield of protons of range 28 cm (±1 cm) is found. The yield is about seven times as great as found from the group of energy change +5 Mev. The calculated energy change is +2.39±0.10 Mev on the assumption that the reaction responsible is the same as for the previous groups, namely $B^{10}+D^2 \rightarrow B^{11}+H^1$.

The great yield of this group suggests that it could be due to $B^{11}+D^2 \rightarrow B^{12}+H^1$ for which the energy change would be 2.38 Mev and the resulting mass for B^{12} would be 12.0170. This would agree very well with the inspection upper limit for the electrons from B^{12} as found by Crane[6] and the amount of the yield is about right. The work of Smith and Murrell[7] in which the separated isotopes of boron were studied showed no group corresponding to this one in the B^{11} target. Therefore the reaction involving B^{12} would appear to be ruled out. The absence of such a group in their experiments is still hard to explain. It has been suggested that a gamma-ray of approximately 2 Mev accompanies the electrons which would make the energy change for this reaction about 0.4 Mev. We have looked for such a group but it appears to fall almost coincident with the very prolific alpha-particles from the reaction $B^{10}+D^2 \rightarrow Be^8+He^4$ and we cannot say that it is present.

In Fig. 3 the excited states of B^{11} found from

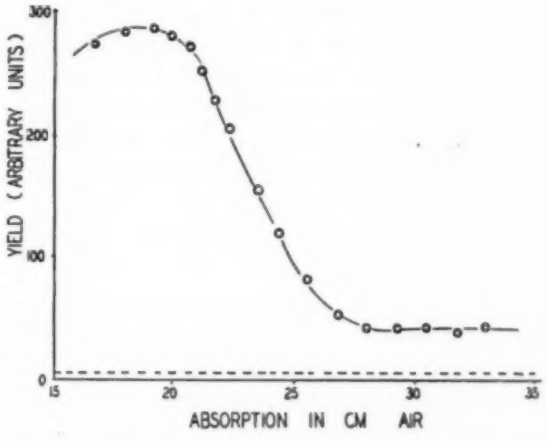

FIG. 2. Absorption curve for the new group of protons. The yield is sufficiently large to fit the reaction $B^{11}(dp)B^{12}$ but the absence of a corresponding group in the work of Smith and Murrell on the separated isotopes of boron indicates that it is probably due to a third excited state of B^{11}.

[4] D. W. Kerst, Rev. Sci. Inst. **9**, 131 (1938).
[5] S. K. Allison, Phys. Rev. **55**, 624 (1939).
[6] D. S. Bayley and H. R. Crane, Phys. Rev. **51**, 1012 (1937).
[7] C. L. Smith and E. B. M. Murrell, Proc. Camb. Phil. Soc. **35**, 298 (1939).

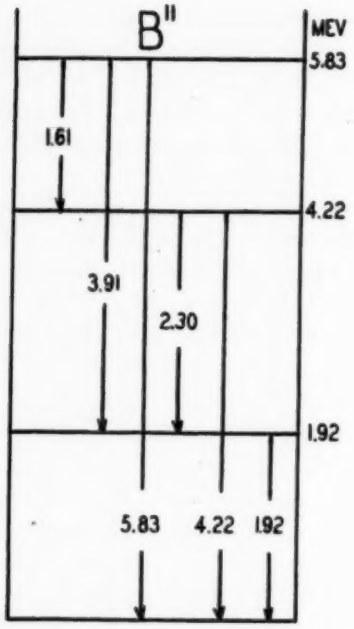

FIG. 3. Energy level diagram for B^{11}. The energies corresponding to the possible optical transitions are given.

this work are represented on a diagram. It will be seen that the spacing is roughly uniform. Gamma-rays as indicated may be expected. The gamma-rays from boron bombarded by deuterons have been studied by Gaerttner, Fowler and Lauritsen[8] who find gamma-rays of energy 1.5, 2.2, 4.4, 6.9, 9.1 Mev with intensities >2.5, 2.5, 1.0, 0.3, 0.1, respectively. They are able to account for all these except the 1.5-Mev line, on the assumption that the two reactions $B^{10}+H^2 \rightarrow B^{11}+H^1$ and $B^{11}+H^2 \rightarrow C^{12}+n$ are responsible. Our results indicate that the 1.5-Mev line can be explained partly as a transition from the third to the second excited state of B^{11}. There must be some other transition producing a line of about this energy as the above explanation would require large intensity for the subsequent transitions also. The 4.4- and 2.2-Mev lines are due in part to excited states of C^{12} and a large intensity cannot be assigned to B^{11}. It is therefore likely that the 1.5-Mev line is in part due to a transition following the decay of B^{12} to C^{12}. Proof of this is not easy as B^{12} has a half-life of 1/50 second.

The relative yields of the groups we observe to be roughly 1 : 1.5 : 6 : 28 starting with the group of maximum range. It is of interest to compare this with the values 0.4 : 0.13 : 0.47 for the three outer groups given by Cockcroft and Lewis. It can be seen that with increasing excitation of the compound nucleus caused by the more energetic deuterons, the probability of a transition to an excited state increases. This is to be expected since the transition to the ground state is proportional to the reciprocal of the total number of levels below the total excitation energy. Increase in the number of levels will diminish the relative yield of the maximum energy group as is actually observed.

In conclusion we wish to express our thanks to Professor E. O. Lawrence, Dr. Donald Cooksey, and their associates at Berkeley, who have so generously helped with information and suggestions in addition to supplying the acceleration chamber we use. We wish also to thank Professor Norman I. Adams for invaluable help in designing the R.F. system. A grant from the George Sheffield Fund towards the construction cost is gratefully acknowledged.

[8] E. R. Gaerttner, W. A. Fowler and C. C. Lauritsen, Phys. Rev. **55**, 27 (1939).

Radiations from Radioactive Indium (116)[1]

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(Received April 8, 1940)

The energy of the gamma-radiation emitted by indium (116) with the 54-minute period has been studied with a large hydrogen-filled cloud chamber. The indium was activated by slow neutrons from a cyclotron. The investigation used both the Compton recoil electrons from a carbon radiator of 35 mg/cm² surface density and the photoelectrons ejected from a lead sheet of 20 mg/cm² surface density. The recoil electrons from the carbon radiator indicate that the gamma-radiation is complex and that the spectrum can be resolved into three lines of 1.02, 1.40, and 1.85±0.05 Mev energy, the relative intensities of these lines being 1, 1, and ¼, respectively. The momentum distribution of the photoelectrons emitted from the lead radiator indicates that there are three low energy gamma-ray lines. These lines have energies of 0.17, 0.36, and 0.57±0.03 Mev and each has a relative intensity of about 1/7 compared with the 1.02-Mev line. The beta-ray spectrum of the 54-minute period has been investigated and the upper limit of the energy found to be 0.84±0.01 Mev. A measurement of the ratio of the beta-rays to the gamma-rays shows definitely that more than one quantum is emitted per electron. A level scheme is proposed which is in good agreement with all the data.

INTRODUCTION

INDIUM has two stable isotopes of masses 113 and 115. When it is bombarded with slow neutrons there are two strong activities formed, one of 13 seconds and one of 54 minutes half-life, as first reported by Amaldi, d'Agostino, and Segrè.[2,3] These two periods have been shown by various investigators[4,5] to be due to an isomer of indium (116). In the present paper the gamma-radiations associated with the 54-minute period will be reported.

APPARATUS

These gamma-radiations were studied by means of both the Compton recoil electrons and the photoelectrons emitted from radiators placed across a large cloud chamber 30 cm in diameter. The large diameter of the chamber made possible accurate measurements of the curvatures of the electron tracks. The chamber was traversed by a magnetic field which was constant to ±0.5 percent throughout the region in which the tracks were measured. The magnetic field was kept on continuously, thus eliminating errors due to fluctuations in the field caused by circuit switches and the inaccuracy in reading the ballistic swing of the ammeter.

The radiator used for the Compton recoil tracks was made of paper covered with paraffin and lampblack. It had a surface density of 35 mg/cm². The photoelectrons were ejected from a thin lead sheet of 20 mg/cm² surface density. The cloud chamber was filled with hydrogen at atmospheric pressure, and a mixture of ethyl alcohol and water was used for the condensible vapor. The use of hydrogen is essential when working with particles of the energy considered here in order to reduce the scattering of tracks in the gas of the chamber.

The cloud chamber was illuminated by three 110-volt, 500-watt lamps flashed on 200 volts d.c. The vacuum tube control circuit has been described by Richardson.[6]

The gamma-ray source was placed at one end of a lead collimator approximately 20 cm outside the chamber wall. This was arranged in such a way that gamma-rays could not hit the floor or the roof of the cloud chamber unless scattered several times, thus reducing the number of tracks coming from the material surrounding the chamber.

RESULTS

The radioactive sources of indium (116) were prepared by activating samples of indium with

[1] A preliminary report of this work was given before the American Physical Society, Washington Meeting, 1938.
* Now at Harvard University.
† National Research Fellow. Now at the University of Illinois.
[2] E. Amaldi, O. d'Agostino and E. Segrè, Ricerca Scient. 5, 2 (1934).
[3] E. Amaldi, O. d'Agostino, E. Fermi, B. Pontecorvo, F. Rasetti and E. Segrè, Proc. Roy. Soc. 149, 522 (1935).
[4] L. Szillard and T. A. Chalmers, Nature 135, 98 (1935).
[5] J. L. Lawson and J. M. Cork, Phys. Rev. 52, 531 (1937).
[6] J. R. Richardson, Rev. Sci. Inst. 9, 152 (1938).

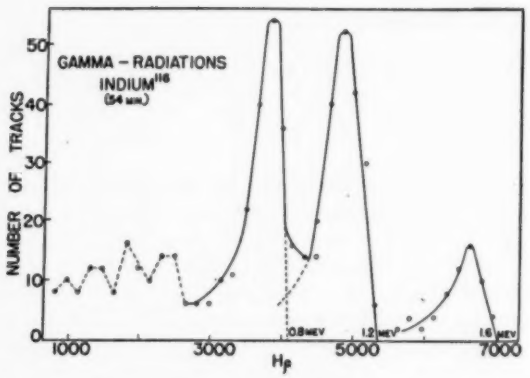

FIG. 1. The momentum distribution of the Compton recoil electrons from a carbon radiator of 35 mg/cm² surface density. Three gamma-ray lines are present of 1.02, 1.40, and 1.85 Mev energy, the relative intensities being 1, 1, and ¼, respectively.

slow neutrons obtained from the Michigan cyclotron by bombarding beryllium with deuterons. The samples were exposed for one hour, longer bombardments being avoided for fear of building up the intensity of any gamma-radiations that might be due to longer period indium activities. The sources were allowed to decay a sufficient length of time to permit the 13-second period to disappear before the pictures were taken with the cloud chamber.

A. Compton electrons

The momentum distribution of the Compton recoil electrons is shown in Fig. 1. There are clearly three main groups which are quite well resolved. The upper limits of the Compton recoil electrons occur at 4100, 5350, and 7000$H\rho$, corresponding to energies of 0.82, 1.18, and 1.63 Mev, respectively. When the energy of the recoil quantum is included, the three lines are found to be due to gamma-rays of 1.02, 1.40, and 1.85 Mev energy. The intensities of the three lines are 1, 1, and ¼, respectively. The limit of accuracy on the measurements may be set as ± 0.05 Mev for each line. The fact that part of the radiation is over 1.65 Mev has also been verified by observation of the photodisintegration of beryllium by this radiation.

B. Photoelectrons

Two lines of lower energy were included in our previous report. This lower energy region has been reinvestigated by obtaining a more complete distribution of the photoelectrons ejected from a lead radiator of 20 mg/cm² surface density. The results are shown in Fig. 2. The question as to whether or not these data can be represented by a smooth monotonic curve may be considered. One can estimate fairly well from the standard deviations involved that there is approximately one chance in ten that such is the case. With this reservation, then, one can feel fairly sure that the indicated peaks are real.

The peak observed at 1870$H\rho$, for example, because of its intensity is clearly composed of electrons from the K shell. This momentum must be corrected for the shift in the peak of the photoelectron curves due to the loss of energy of the particles in passing through the lead radiator. Some idea of the amount of this shift can be obtained from the data of White and Millington[7] (corrected for the difference in atomic number and the fact that photoelectrons may start anywhere in the radiator). The shift in the peak for this particular momentum and radiator thickness will be approximately 100$H\rho$. Thus the corrected value for the momentum of the photoelectrons should be 1970$H\rho$ or an energy of 0.271 Mev. To this must be added the binding energy of the K shell, making the energy of the observed gamma-radiation 0.358 Mev. Because of the scattering of the photoelectrons within the lead, and the possibility of statistical fluctuations, the accuracy of the line is set at ± 0.03 Mev.

The most prominent peak at 850$H\rho$ is of such low energy that not all of the photoelectrons will get out of the radiator. The sharp decrease to zero on the low energy side of this peak is to be expected even though this is true, because of the way in which the energy loss of electrons varies with the velocity. In accord with this fact, and with the assumption that the electrons are from the K shell, one finds that the energy of the gamma-ray must be 0.17 ± 0.03 Mev. The peak at 1320$H\rho$ is of just the right energy and relative intensity to be composed of electrons from the L shell ejected by the gamma-ray of 0.17 Mev. There is also evidence of a third line with an observed momentum of 2760$H\rho$ which after correction gives an energy of 0.57 ± 0.03 Mev.

[7] P. White and G. Millington, Proc. Roy. Soc. **A120**, 701 (1928).

The intensity of each of these lines has been determined from the values for the photoelectric absorption cross section given by Heitler.[8] The three low energy lines are all of about equal intensity. The accuracy of the intensity measurements is not very high and they may be in error by a factor of two.

A comparison of the intensities of these lines with those of the Compton lines can be obtained from the number of tracks above $3000H\rho$, which are due to the Compton recoils and photoelectrons emitted by the lines of 1.0, 1.4, and 1.8 Mev energy. Assuming that these tracks are mainly due to the two strongest lines of 1.0 and 1.4 Mev energy, the intensity of the 0.19-, 0.36- and 0.57-Mev lines is approximately 1/7 for each.

C. The beta-ray spectrum

The beta-ray spectra from the 13-second and 54-minute periods of indium (116) have been investigated by several groups. Gaerttner, Turin and Crane,[9] using a cloud chamber, found an upper limit of the 54-minute period spectrum to be 1.3 Mev, while Brown and Mitchell[10] report an extrapolated Konopinski-Uhlenbeck plot to give an endpoint of 1.45 Mev.

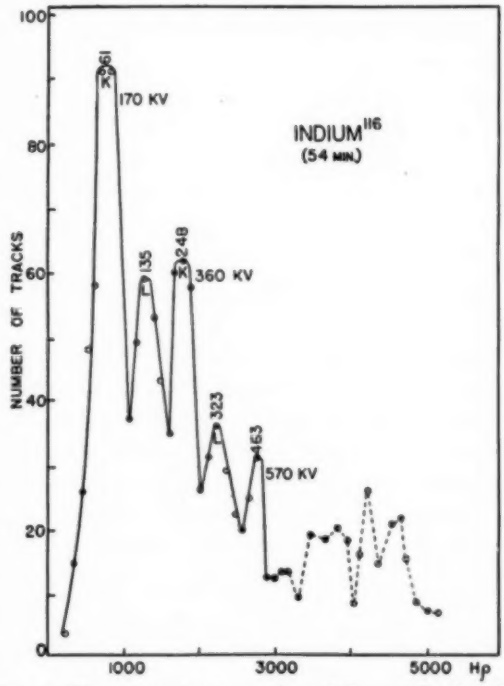

FIG. 2. The photoelectrons ejected from a lead radiator of 20 mg/cm² surface density by the gamma-radiation from the 54-minute period of indium (116). Three gamma-ray lines are indicated with energies of 0.17, 0.36 and 0.57 Mev. The relative intensities of the three lines are approximately equal.

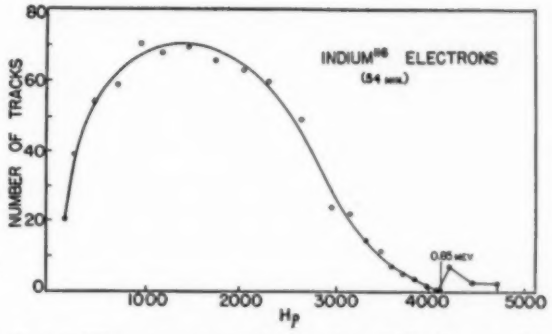

FIG. 3. The momentum distribution of electrons emitted by the 54-minute isomer of indium (116). The observed upper limit is 0.84 Mev energy. The slight peak at 4300 $H\rho$ is due to the gamma-rays of 1.4 Mev.

Later work by both cloud chamber and magnetic spectrometer show that this endpoint is quite in error. In the present investigation the beta-ray spectrum was again studied by the cloud-chamber method and the momentum distribution is shown in Fig. 3. The upper limit by inspection occurs at $4150H\rho$ or an energy of 0.84 Mev, which is in good agreement with the spectrometer[11] value of 0.85 ± 0.01 Mev. The slight peak observed at $4300H\rho$ is probably due to Compton electrons formed in the source from the 1.4- and 1.8-Mev gamma-ray lines.

The explanation for the discrepancy between the previous work and the later investigations is that the earlier experiments were done with slow neutrons from radon-beryllium sources and in order to gain sufficient intensity, thick pieces of indium were used. This would cause Compton electrons to be emitted from the source itself, and as the present work indicates that there are gamma-rays of 1.0 and 1.4 Mev energy present, it is not surprising that the endpoint for the spectrum was increased to nearly 1.4 Mev energy. In the new determinations of the endpoint, very thin indium foils were used and the error due to the gamma-ray lines is reduced.

The beta-ray spectrum for the 13-second period has not been remeasured and the value for the upper limit is accepted as that of 3.1 Mev

[8] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, 1935), p. 126.

[9] E. R. Gaerttner, J. J. Turin and H. R. Crane, Phys. Rev. **49**, 793 (1936).

[10] M. V. Brown and A. C. G. Mitchell, Phys. Rev. **50**, 593 (1936).

[11] J. M. Cork and J. L. Lawson, Phys. Rev. **56**, 291 (1939).

found by Gaerttner, Turin and Crane.[9] Absorption measurements[12] indicate that the endpoint of the spectrum has a value of approximately 2.8 Mev, which would indicate that the cloud-chamber measurements were too high.

D. Energy level system for the isomers of indium (116)

There is now sufficient data to make an energy level diagram for the two isomers assigned to radioactive indium (116). Although the nuclear energy diagram may be revised when more accurate methods are found for measuring the gamma-ray energies, the present information is sufficient to give an idea of the existing transitions. A suggested nuclear energy arrangement is shown in Fig. 4. The nucleus of indium (115) is lifted up to some excited level by the addition of the neutron, and after the emission of several gamma-rays the two isomers of indium (116) exist at some excited level approximately 3.1 Mev above the ground state. They may then lose energy to reach the ground level in several different ways.

The 13-second period has been investigated by Langer and Mitchell,[13] who report that there are no gamma-rays connected with this isomer. Therefore it must reach the ground state by the emission of a beta-particle plus a neutrino to get rid of 3.1 Mev energy. Langer, Mitchell and McDaniel[14] have studied the gamma-rays from the 54-minute period by means of the coincidence counter method. Their curves show a definite break at 0.8 and 1.2 Mev. It is interesting to note that these breaks occur at two values of the Compton recoil electrons (excluding the energy of the recoil quantum) recorded in this paper. They report that the 54-minute period has two gamma-rays connected with it.

The evidence presented in this investigation shows that the 54-minute period can reach the ground state by several different transitions. After the emission of a beta-particle plus a neutrino of 0.85 ± 0.01 Mev energy to tin (116), the rest of the energy may be lost by any of three gamma-ray transitions. It may lose energy by a 1.40 ± 0.05-Mev gamma-ray followed by a

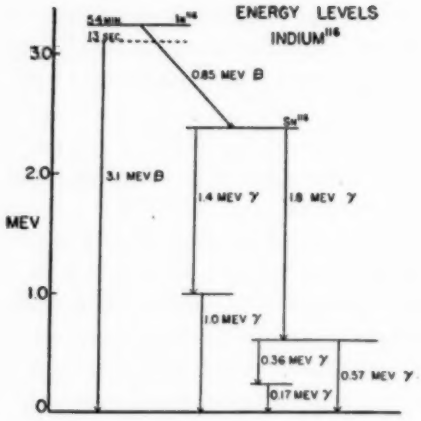

FIG. 4. A suggested nuclear energy level diagram for the 13-second and 54-minute isomers of indium (116). The gamma-ray transition of 1.0 and 1.4 Mev energy has a probability of 0.75 and the transition by the 1.8-Mev energy gamma-ray has a probability of 0.25.

1.02 ± 0.05-Mev line. This process has a probability of 0.75. The sum of the beta-ray transition plus the gamma-rays is 3.27 ± 0.08 Mev. Another possible transition is by the emission of a 1.85 ± 0.05-Mev gamma-ray followed by gamma-rays of 0.36 ± 0.03 Mev and 0.17 ± 0.03 Mev energy. The sum of the energies of this transition is 3.23 ± 0.07 Mev. The 1.85-Mev gamma-ray has a probability of $\frac{1}{4}$, whereas the lower energy lines have a probability of 1/7. This would indicate that there is another line of intensity 0.1 with an energy equal to the sum of the two low energy lines. The 0.57 ± 0.03-Mev gamma-ray fulfills the necessary condition as far as energy is concerned, but its intensity is about the same as that of the 0.17- and 0.36-Mev gamma-rays. The intensity measurements are not very accurate, and it is quite possible that if more data were acquired the intensities of these lines would be somewhat different. The sum of the energies of the final transition path is 3.27 ± 0.06 Mev. The fact that there is no gamma-radiation corresponding to the full transition, $1.0+1.4=2.4$ Mev, fits in quite nicely with the idea that the spin change must be large in these transitions.

All the transitions seem to indicate, within the limits of experimental error, that the 54-minute isomer exists at a higher level than the 13-second isomer, assuming that the 13-second beta-ray spectrum has been accurately measured.

This work has been made possible by a grant from the Horace H. Rackham Fund.

[12] J. L. Lawson, private communication.
[13] A. C. G. Mitchell and L. M. Langer, Phys. Rev. **53**, 505 (1938).
[14] L. M. Langer, A. C. G. Mitchell and P. M. McDaniel, Phys. Rev. **56**, 380 (1939).

A Continuum Theory of the Compound Nucleus

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(Received March 21, 1940)

The nucleus is described by an absorption coefficient σ which gives the probability per unit time that an incident particle becomes amalgamated with the nucleus (Eq. (1)). This absorption coefficient appears as an imaginary potential in the Schrödinger equation. It is shown that a gradual decrease of σ at the nuclear boundary is essential for achieving agreement with experiments (§2). This model gives automatically unit sticking probability for fast neutrons, a cross section proportional to $1/v$ for slow neutrons, and no one-particle resonances for particles which have to penetrate a potential barrier (§3). Quantitative calculations are made with σ varying as $e^{-(r-R)/b}$ outside the nucleus. For neutrons of zero orbital momentum, the formation probability of the compound nucleus is found to be $\zeta=1-e^{-2\pi kb}$ where k is the wave number. It is significant that ζ depends on the diffuseness b of the nuclear boundary rather than on the nuclear radius R. On the other hand, the factor 2π ensures that ζ is close to unity already for energies of about 1 Mev (§4). The total cross section in the region of overlapping levels, and the average level width in the region of separated levels are expressed in terms of the formation probability ζ. The relation with the elastic scattering is discussed (§5). The case of slow neutrons is treated in detail. With an average spacing D of 10 volts between levels of the same J, the average neutron width is about $2\times10^{-3}E^{\frac{1}{2}}$ for a neutron energy E, in rough agreement with the meager experimental data. With these assumptions, the neutron width will become larger than the radiation width already for $E\approx10^8$ ev; experiments on the capture of "medium fast" neutrons ($\approx2\times10^5$ ev) can be interpreted roughly on this basis. The elastic potential scattering of slow neutrons is shown to be equivalent to the scattering from a hard sphere whose radius R' is defined by the condition that $\sigma(R')=(\hbar/2mb^2)e^{-2C}$ where C is Euler's constant $0.577\cdots$ (§6). The case of particles which move in a non-nuclear potential V (electrostatic or centrifugal) is treated in §4, 7, 8 for various relations between the energy E of the incident particle and the height $V(R')$ of the potential barrier. If $E-V(R')$ is more than about 1 Mev, the formation probability is close to one, as for a fast neutron (§4). If E is about equal to $V(R')$, ζ is still of the order of unity (§8). For $E<V(R')$, ζ contains the well-known penetrability of the potential barrier, e^{-2G}, aside from other factors which increase slowly with $|E-V(R')|$ (§7). The magnitude of σ inside the nucleus is derived for the case of extremely high energies from the Born approximation and the variation of σ with energy is shown to be slight in this case. Although quantitative conclusions on the case of moderate energies cannot be drawn, it seems likely that σ is at least 20–40 Mev in that case (§9). Finally, it is shown that no appreciable change of results is caused by an attractive or repulsive nuclear potential added to the nuclear absorption potential (§10). In the main part of the paper, it has been assumed that the average interaction between nucleus and particle is zero.

§1. INTRODUCTION

THE treatment of the compound nucleus has in the past been based mostly on dispersion theory.[1–5] This treatment is eminently well suited for the resonance phenomena observed with slow neutrons and protons for which it was originally devised by Breit and Wigner.[1] Moreover, Bohr, Peierls and Placzek[5] have shown that the dispersion theory (in the form of Peierls and Kapur[4]) can also be applied in the region of dense levels, i.e. when the width of the compound levels is greater than their spacing. On the other hand, it is of course *not necessary* to use dispersion theory in the region of overlapping levels because the cross section then no longer depends on the individual energy levels. It is instead permissible to treat the energy levels statistically which means a great simplification of the formalism.[6–8] Bohr, Peierls and Placzek have shown that the statistical and the dispersion method give the same cross section for the formation of the compound nucleus and have investigated which quantities in dispersion theory correspond to the quantities used in the statistical treatment.

In either case, quantitative results can only be obtained when certain assumptions are made

[1] G. Breit and E. Wigner, Phys. Rev. **49**, 519 (1936).
[2] H. A. Bethe and G. Placzek, Phys. Rev. **51**, 450 (1937).
[3] N. Bohr and F. Kalckar, Kgl. Dansk. Vid. Selsk., Math.-fys. Med. **14**, 10 (1937).
[4] P. L. Kapur and R. Peierls, Proc. Roy. Soc. **166**, 277 (1938).
[5] N. Bohr, R. Peierls and G. Placzek, to appear shortly. I am indebted to Dr. Placzek for the opportunity to see the manuscript before publication.
[6] L. Landau, Physik. Zeits. Sowjetunion **11**, 556 (1937).
[7] V. F. Weisskopf, Phys. Rev. **52**, 295 (1937).
[8] V. F. Weisskopf and D. H. Ewing, Phys. Rev. **57**, 472 (1940).

about the physical quantities entering the theory. In the statistical theory it is shown that the total cross section can be expressed in terms of the sticking probability ξ, i.e., the probability that an incident particle hitting the nucleus will stick to it and thus form a compound nucleus. It is usually assumed that this sticking probability is unity for high energy particles and proportional to the velocity for slow particles, but it has not been possible thus far to give a general theory comprising both limiting cases and permitting an interpolation between them, or even to say which energies are to be considered as "high" and "low."

In this paper, we shall attempt to give a theory of the sticking probability. For this purpose, we shall consider the case of dense levels and shall try to develop a *continuum theory* of the compound nucleus, disregarding all effects of the individual levels. This theory will be related to the dispersion theory as the classical theory of a solid—describing it by phenomenological constants like conductivity or elasticity—is related to a quantum theory which would take into account each (electronic and vibrational) quantum state of the crystal.

In our continuum theory, we shall start from the basic idea underlying the theory of the compound nucleus,[9] *viz.* that there is a strong interaction between a nucleus and any nuclear particle incident upon it which leads to an amalgamation of the two. This means that there is a large probability for a "free" nuclear particle inside a nucleus to become absorbed and lose its individuality. We shall therefore *describe a nucleus by a large absorption coefficient for nuclear particles*. For reasons to be explained later (below Eq. (2a)), we shall refer to this absorption coefficient as "absorption potential."

An incident particle can then either be absorbed by the initial nucleus or elastically reflected. Our aim will be to calculate the relative probabilities of these two processes (in other words, the sticking probability) in terms of the absorption potential and of the velocity of the incident particle. The absorption of the particle is, of course, equivalent to the formation of the compound nucleus, and therefore the absorption probability gives the total cross section for all possible reactions. How this cross section is distributed over the various possible reactions is another matter; according to the original hypothesis of the compound nucleus, its disintegration is independent of the manner of its formation, and, if this is true, the relative probabilities of various modes of disintegration can be calculated by statistical arguments from the probabilities of the inverse processes.[8]

[9] N. Bohr, Nature **137**, 344 (1936).

§2. Definition of the Absorption Potential

We have proposed in the preceding section to describe the nucleus by an absorption coefficient. We shall assume that the number of particles absorbed per unit time at a given point in the nucleus is proportional to the density of particles at that point. Then we have the continuity equation

$$\partial\rho/\partial t+\text{div}\,\mathbf{j}=-2\sigma(\mathbf{r})\rho/\hbar, \qquad (1)$$

where $\mathbf{j}$ is the current density, ρ the density of particles and σ the "absorption potential." The function σ is defined so as to have the dimension of an energy; it will in general depend on the position $\mathbf{r}$ in the nucleus.

Equation (1) contains the assumption that the absorption depends only on the density of the particles but not on their kinetic energy, direction of motion, spin, etc. This assumption cannot be justified from first principles. However, it seems reasonable to assume that the dependence on spin and direction is slight, and that the dependence on the energy E is only appreciable if E changes at least by an amount of the order of a nuclear binding energy ($\sim$10 Mev). That the dependence on E is slight for very high energies E, will be shown in §9. Then, for our purposes, σ may be considered as a function of $\mathbf{r}$ only.

If we use the wave-mechanical definition of density ρ and current density $\mathbf{j}$, we can show easily that (1) is equivalent to the Schrödinger equation

$$\nabla^2\psi+(2m/\hbar^2)(E-V+i\sigma)\psi=0, \qquad (2)$$

where E is the energy of the incident particle and V its ordinary (real) potential energy. In order to show that (2) is equivalent to (1), we multiply (2) by ψ^* and subtract from the product its

complex conjugate; remembering then the definitions of density and current density, *viz.*,

$$\rho=\psi^*\psi,\ \mathbf{j}=(\hbar/2im)(\psi^*\ \text{grad}\ \psi-\psi\ \text{grad}\ \psi^*),\quad (2a)$$

we obtain Eq. (1) after a simple transformation. In Eq. (2), σ appears as the imaginary part of the potential. This justifies our use of the term "absorption potential," in preference to the colorless term "absorption coefficient."

The absorption potential σ may be assumed to be a function only of the distance r from the center of the nucleus. Inside the nucleus $(r<R)\sigma$ will be of the order of the nuclear interaction forces, i.e., perhaps 30 Mev (cf. §9). At the nuclear boundary σ will drop to zero. We shall show presently that the results for the sticking probability will depend most sensitively on the way in which σ goes to zero, i.e., whether this happens suddenly or gradually. On the other hand, the results will be practically independent of the absolute magnitude of σ inside the nucleus. (§3, 9).

If our theory is to agree with experimental results, it must give a sticking probability very close to unity for fast neutrons ($\gg$1 Mev). This follows from experiments of Bacher[10] on the scattering of fast neutrons by Cu and Pb. These experiments show that practically all the scattering is inelastic; what little elastic scattering there is, goes mainly in the forward direction[10,11] and is readily explained by shadow diffraction.[5, 11a] Therefore experimentally the nucleus behaves like a blackbody to incident fast neutrons; it absorbs practically all of the incident particles and reflects only very few elastically.

In order to find out how σ must behave in order to give blackbody properties to the nucleus it is convenient to remember the electrodynamic analog. Consider a medium of complex refractive index $N=n+ik$ with a sharp plane boundary. If light falls normally on the boundary from the vacuum, the reflection coefficient is

$$R=\left|\frac{N-1}{N+1}\right|^2=\frac{(n-1)^2+k^2}{(n+1)^2+k^2}.\quad (3)$$

All the light which is not reflected is absorbed if the medium is sufficiently thick; the fraction absorbed is equivalent to the sticking probability in nuclear physics and (for infinite thickness) is given by

$$\zeta=1-R=4n/[(n+1)^2+k^2].\quad (3a)$$

From this formula it is seen that a large absorption coefficient k is by no means favorable for a large sticking probability; on the contrary, the only case when ζ becomes nearly 1 is for $N=n+ik$ nearly 1, i.e. when the medium is very similar to vacuum. We know, however, that the complex refractive index N is by no means close to unity in the nuclear case; in fact, $|N^2-1|=|-V+i\sigma|/E$ is in general large compared with unity. Therefore, if the nucleus had a sharp boundary, we would obtain a large reflectivity and small sticking probability.

The result is quite different if σ and V change *gradually* at the nuclear boundary. If the variation of $U=E-V+i\sigma$ over one wave-length is small compared with U itself, we can use the WKB method. As is well known, this method is equivalent to classical mechanics; therefore a stream of particles incident upon the nucleus will proceed towards the nucleus without being reflected, until they arrive in the regions where they are absorbed. Therefore a model in which the complex potential $V-i\sigma$ changes gradually at the nuclear boundary will, for fast incident particles (small wave-length), give *nearly unit sticking probability*.

A gradual variation of V and σ is very plausible. Both these quantities are connected with the forces between nuclear particles. Therefore, even if the nucleus had a sharp boundary, V and σ would extend beyond the geometrical boundary over a distance of the order of the range of the nuclear forces, i.e., about 10^{-13} cm. Actually, the boundary of the nucleus itself will not be sharp, because of the zero-point oscillation of the nuclear particles, which constitutes a further reason for a gradual change of V and σ. We shall use the term "diffuseness of the nuclear boundary," b, for the distance over which the absorption potential σ (and also V) fall to, say, $1/e$ of their value inside the nucleus. According to the foregoing, there are two contributions to

[10] R. F. Bacher, Phys. Rev. **55**, 679 (1939); **57**, 352 (1940).
[11] T. Wakatuki and S. Kikuchi, Proc. Phys.-Math. Soc. Japan **21**, 656 (1939).
[11a] G. Placzek and H. A. Bethe, Phys. Rev. **57**, 1075A (1940).

b, probably of about equal importance, one being the diffuseness of the region actually occupied by the particles in the nucleus and the other the range of the nuclear forces.

We thus see that very plausible assumptions lead to unit sticking probability for fast neutrons. The same assumptions will also eliminate one-particle resonances without the artificial introduction of a repulsive potential.[2] As we shall see in §3, our absorption theory will give the "$1/v$ law" for slow neutrons and Weisskopf's formula[8] for charged particle reactions.

§3. General Calculation

Many results can be obtained without special assumptions about the potential V and the absorption potential σ. The potential V will in general contain one part, V_n, which arises from nuclear forces, and another part, V_{ext}, which is not of nuclear origin and contains the Coulomb and the centrifugal force. Since the diffuseness of the nuclear boundary (cf. end of §2) is small, we can for most purposes consider the slowly varying part V_{ext} as constant over the interesting region, i.e. over the region outside the nucleus in which V_n and σ are still appreciable (cf., however, §8). We shall introduce

$$W=E-V_{ext}(R), \tag{4}$$

i.e., the kinetic energy of the incident particle at the surface of the nucleus (R=nuclear radius) disregarding nuclear forces. Then the radial Schrödinger equation is

$$\frac{d^2u}{dr^2}+\frac{2m}{\hbar^2}(W-V_n+i\sigma)u=0, \tag{5}$$

where u is r times the radial factor of ψ. We shall frequently use the abbreviation

$$\Phi=(2m/\hbar^2)(-W+V_n-i\sigma). \tag{5a}$$

A. High energy

If W is large and positive, the WKB method will be applicable for all values of r. Then

$$u=c\Phi^{-\frac{1}{4}}\exp\left(\int\Phi^{\frac{1}{2}}dr\right), \tag{6}$$

where c is a constant. The square root of Φ has to be chosen in such a way that the wave function (6) *decreases* towards the interior of the nucleus because the nucleus absorbs particles coming from the outside but does not emit any. This means that the real part of $\Phi^{\frac{1}{2}}$ must be positive. Now the imaginary part of Φ (cf. (5a)) is negative; therefore $\Phi^{\frac{1}{2}}$ must lie in the fourth quadrant of the complex plane (real part positive, imaginary part negative). As σ and V_n decrease, Φ becomes real and negative; and, since $\Phi^{\frac{1}{2}}$ stays all the time in the fourth quadrant, it will become purely negative imaginary far outside the nucleus. Therefore (6) goes over automatically into

$$u=c\Phi^{-\frac{1}{4}}\exp\left(-i(2mW)^{\frac{1}{2}}r/\hbar\right) \tag{6a}$$

or, for still larger distances, into

$$u=c\Phi^{-\frac{1}{4}}\exp\left\{-i(2m)^{\frac{1}{2}}\hbar^{-1}\int(E-V_{ext})^{\frac{1}{2}}dr\right\}, \tag{6b}$$

in other words, into an incident wave without any outgoing part. Therefore, for high energy, the nucleus does not reflect any particles; the sticking probability is

$$\zeta=1. \tag{6c}$$

Any deviation from this result is due to the inaccuracy of the WKB.

B. Small energy

For slow neutrons of zero orbital momentum, we have $V_{ext}=0$ and $W=E$ small. Then the WKB method is valid only inside the nucleus, i.e., as long as $|V_n-i\sigma|$ is sufficiently large. Let R be the value of r for which the WKB method just ceases to be valid. Then at $r=R$ we have approximately, according to (6):

$$\left(\frac{1}{u}\frac{du}{dr}\right)_{r=R}\approx\Phi^{\frac{1}{2}}(R)\approx\left[\frac{2m}{\hbar^2}(V_n-i\sigma)\right]^{\frac{1}{2}}. \tag{7}$$

The condition for validity of the WKB method is that the change of Φ over one wave-length be small compared to Φ itself,[12] one wave-length being equal to $|\Phi^{-\frac{1}{2}}|$. At the limit of validity, then, $|\Phi^{-\frac{1}{2}}|$ will be approximately equal to the

[12] Actually, the quantity neglected in the WKB is only the second derivative of $\Phi^{-\frac{1}{4}}$, rather than of Φ. Therefore the limit of validity may be given by the point where the wave-length $|\Phi|^{-\frac{1}{2}}$ is equal to $4b$ rather than to b. This would increase the result (11) for ζ by another factor 4. (With the exponential potential of §4, ζ comes out about 2.2 times the value given in (11).)

distance over which Φ changes by its own amount, i.e., of the order of the diffuseness of the nuclear boundary, b. Thus we have

$$(d \log u/dr)_R = (1/b)e^{-\frac{1}{2}i\varphi} \tag{7a}$$

with

$$\tan \varphi = (\sigma/V_n)_R. \tag{7b}$$

Immediately outside R, there will be a small region in which V_n and σ are still important but the effect of this region on $d \log u/dr$ is slight. We can therefore join the wave function at $r=R$ immediately to that of a free particle of energy E, *viz.*

$$u = \sin(kr+\delta) \tag{8}$$

with

$$k = (2mE)^{\frac{1}{2}}/\hbar. \tag{8a}$$

Since k and δ are small, we can write for r of the order R:

$$u = kr+\delta, \tag{8b}$$

$$\left(\frac{1}{u}\frac{du}{dr}\right)_R = \frac{k}{kR+\delta}. \tag{8c}$$

Equating (8c) and (7a), we find

$$\delta = -kR+kbe^{\frac{1}{2}i\varphi}. \tag{9}$$

The term kR gives a potential scattering with a cross section $4\pi R^2$. From the other term we may obtain the sticking probability (besides an additional contribution to the scattering). For this purpose, we re-write (8) in the form

$$2iu = e^{ikr+i\delta} - e^{-ikr-i\delta}. \tag{10}$$

This expression consists of an incident wave and a reflected wave, the ratio of their amplitudes being $e^{2i\delta}$. The reflection coefficient is then

$$P = |e^{4i\delta}| = \mathrm{Re}\,(1+4i\delta), \tag{10a}$$

where terms of order δ^2 have been neglected and Re denotes the real part. The sticking probability is then (cf. (9))

$$\zeta = 1-P = \mathrm{Im}\,(4\delta) = 4kb \sin \tfrac{1}{2}\varphi. \tag{11}$$

According to (7b), $\sin \frac{1}{2}\varphi$ will be of the order unity if σ is comparable to V_n at the limit of validity of the WKB. Therefore ζ is of the order of $4kb$.

This shows that ζ decreases as $k \sim E^{\frac{1}{2}}$ for slow neutrons. ζ has been defined here as the ratio of the number of particles absorbed to the number of incident particles with $l=0$. The latter number is π/k^2 for unit total incident current; therefore the absorption cross section is

$$\sigma = (\pi/k^2)\zeta \approx 4\pi b/k, \tag{12}$$

i.e., the well-known $1/v$ law. Our theory does of course not describe the resonance phenomena (cf. §6).

It must be pointed out that our definition of ζ differs from the usual definition[13] of the sticking probability ξ. The latter was made so as to make ξ close to unity for any case, including slow neutrons and particles going through a potential barrier. Our definition of ζ is very much simpler, but of course we cannot, in general, expect it to be unity. In order to avoid confusion, we propose to call ζ the "formation probability," i.e., the probability of formation of the compound nucleus.

According to (11), the formation probability will become of the order unity when $\lambda\!\!\!^{-} = 1/k \approx 4b$. This solves a question of old standing, *viz.* whether it is sufficient to make the wave-length $\lambda\!\!\!^{-}$ smaller than the nuclear radius R in order to make the formation probability almost unity, or whether it is necessary to make $\lambda\!\!\!^{-}$ smaller than the diffuseness b of the nuclear boundary. We see that b rather than R is important which is very plausible because, for an imaginary nucleus of very large dimensions, the size can obviously not be important for its physical properties. On the other hand, b is multiplied by the numerical factor 4 which arises from the $e^{4i\delta}$ in Eq. (10a) for the reflection coefficient; this means that already for rather large values of $\lambda\!\!\!^{-}$ (rather small energies) the formation probability becomes unity, in agreement with experiment.[10] It was mainly in view of the experimental results that it had been believed[13] that R was the critical value for $\lambda\!\!\!^{-}$ below which $\zeta \approx 1$. A more quantitative discussion will be given in §4.

C. Negative energy

If W is negative (potential barrier), $\Phi^{\frac{1}{2}}$ will become positive real outside the nucleus (cf. case A). This means that the wave function will be much smaller at the nuclear boundary than

[13] H. A. Bethe, Rev. Mod. Phys. **9**, 69 (1937), (cf. p. 96).

farther out so that our theory leads automatically to the ordinary result of small penetration through the potential barrier, and also to the usual expressions for the penetrability. Furthermore, the increase of the wave function outwards is obtained quite generally which shows that there are no one-particle resonances. The only condition is that the WKB method is applicable everywhere near the nuclear boundary. This means that *if* the nuclear potential V_n is attractive and becomes greater than $|W|$ at a certain point r_0, then $\sigma(r_0)$ should be large enough to insure the validity of the WKB near r_0. This will be the case if σ is of the same order as V_n or larger which is to be expected on general grounds.

§4. Quantitative Calculation for Exponential Absorption Coefficient

For the purpose of a more detailed discussion, we shall assume that σ depends exponentially on r, *viz.*

$$\sigma=(\hbar^2/2mb^2)e^{-(r-R)/b}. \tag{13}$$

b is the "diffuseness of the nuclear boundary." The factor $\hbar^2/2mb^2$ is chosen for convenience in the calculations. Eq. (13) contains a definition of the nuclear radius R, *viz.*, R is that value of r for which σ has the value $\hbar^2/2mb^2$. If we take for b simply the range of the nuclear forces (cf. §2), and if we determine this range from the theory of the triton,[14] we obtain $b=0.86\times10^{-13}$ cm, and $\hbar^2/2mb^2=28$ Mev, i.e. rather large. The expression (13) becomes very large inside the nucleus ($r<R$) whereas we should expect σ to reach a constant value of perhaps 30 Mev. This does not affect the results perceptibly because the inside value of σ is large enough to make the WKB valid for the transition from the inside to the boundary region, and therefore the behavior of the wave function in the boundary region depends only on the value of σ in that region and is independent of the behavior of σ inside the nucleus.

We shall further assume that V_n is zero. This assumption is probably a good approximation because, as far as we know, nuclear forces are sometimes repulsive, sometimes attractive according to the relative orientation of the spins of the interacting particles, their relative orbital momentum, their charge, etc. The interaction of a complicated nucleus, containing many particles of different spin, etc., with another particle will be very close to zero, at least on the average over the surface of the nucleus. We shall show in §10 that finite V_n gives essentially the same results as $V_n=0$.

The radial Schrödinger equation becomes now

$$\frac{d^2u}{dr^2}+\left\{k^2+\frac{i}{b^2}e^{-(r-R)/b}\right\}u=0. \tag{14}$$

Here k is the wave number of the incident particle near the nucleus, *viz.* [cf. Eq. (4)]

$$k^2=2mW/\hbar^2=(2m/\hbar^2)(E-V_{\text{ext}}(R)) \tag{14a}$$

$$=\frac{2m}{\hbar^2}\left(E-\frac{Zze^2}{R}\right)-\frac{l(l+1)}{R^2}, \tag{14b}$$

where l is the orbital momentum of the incident particle, ze its charge, Ze the charge and R the radius of the nucleus.

Equation (14) may be solved by introducing the independent variable

$$x=e^{-(r-R)/2b}. \tag{15}$$

Then (14) becomes

$$\frac{d^2u}{dx^2}+\frac{1}{x}\frac{du}{dx}+4\left(\frac{k^2b^2}{x^2}+i\right)u=0, \tag{15a}$$

whose solution is

$$u=cH_{2ikb}^{(1)}(2i^{\frac{1}{2}}x)=cH_{2ikb}^{(1)}(\sqrt{2}(1+i)x). \tag{16}$$

The Hankel function $H^{(1)}$, rather than any other type of Bessel function, must be chosen in order to make u zero rather than infinite for large x, i.e. inside the nucleus. c is a constant.

We want to know the asymptotic behavior of u far outside the nucleus ($r\gg R$). According to (15), this corresponds to *small* values of x. Neglecting all higher powers of z, we have for small z and any p[15]

$$H_p^{(1)}(2z)=\frac{i}{\sin\pi p}\left(e^{-\pi ip}\frac{z^p}{p!}-\frac{z^{-p}}{(-p)!}\right). \tag{16a}$$

[14] W. Rarita and R. D. Present, Phys. Rev. **51**, 788 (1937).

[15] Cf. Jahnke-Emde, *Table of Functions*, second edition, p. 200 and 194.

Therefore, choosing c appropriately, (16) becomes for small x

$$u=x^p-e^{\frac{1}{2}\pi i p}x^{-p}p!/(-p)! \qquad (16b)$$

Inserting $p=2ikb$ and x from (15), this gives

$$u=e^{-ik(r-R)}-e^{ik(r-R)}e^{-\pi kb+2i\eta}, \qquad (17)$$

where η is the complex phase of $(2ikb)!$, i.e.

$$e^{i\eta}=(2ikb)!/|(2ikb)!|. \qquad (17a)$$

Equation (17) contains the main result. The radial wave function u consists of an incident wave of unit amplitude and an outgoing wave of absolute amplitude $e^{-\pi kb}$. This shows that the amplitude of the outgoing wave is always less than that of the incident one, as it must be. The reflection coefficient of the nucleus is

$$R=e^{-2\pi kb}. \qquad (17b)$$

The difference of the intensities of incident and outgoing wave represents the particles absorbed by the nucleus; the formation probability of the compound nucleus is therefore

$$\zeta=1-e^{-2\pi kb}. \qquad (18)$$

Equation (18) represents the principal result of our theory. It gives the desired expression for the formation probability as a function of the energy of the particle, which is necessary in order to deduce quantitative results for the cross sections of nuclear reactions. The expression (18) has the following properties:

(a) For high energies ($kb\gg 1/2\pi$), the formation probability is practically unity, in agreement with the experiments on fast neutrons. This shows that our assumptions about the absorption potential were appropriate to obtain the desired result.

(b) For low energies ($kb\ll\frac{1}{2}\pi$), the formation probability is

$$\zeta\approx 2\pi kb, \qquad (18a)$$

i.e., ζ is proportional to k and therefore to the velocity. This gives the $1/v$ law for the capture cross section [cf. Eq. (27a)].

(c) The critical wave-length is

$$\lambda\!\!\!^{-}_{cr}=1/k_{cr}=2\pi b. \qquad (19)$$

For a range of the nuclear forces $b=0.86\times 10^{-13}$ cm, we have $\lambda\!\!\!^{-}_{cr}=5.4\times 10^{-13}$ cm. This means that $\lambda\!\!\!^{-}_{cr}$ is of the order of the radius of medium weight nuclei although it is determined not by the nuclear radius but by the diffuseness of the nuclear boundary. Thus our explicit calculation confirms the results obtained in §3 from the WKB. (It should be noted that $\lambda\!\!\!^{-}$ is the ordinary wave-length divided by 2π, so that the "ordinary" critical wave-length would be $4\pi^2 b$.) The energy of a neutron or proton of wave-length $\lambda\!\!\!^{-}_{cr}$ is $E_{cr}=0.7$ Mev. Therefore neutrons above 1 Mev have a sticking probability close to unity. The sticking probability (18a) for slow neutrons may be written

$$\zeta\approx(E/E_{cr})^{\frac{1}{2}} \quad (E\ll E_{cr}). \qquad (19a)$$

According to (14a), k depends on the kinetic energy W of the incident particle at the surface of the nucleus. Only in the case of neutrons with zero orbital momentum, W is identical with the actual energy E and only in this case, Eqs. (18a) to (19a) are directly applicable. In all other cases, W is less than E because of the electrostatic and the centrifugal potential barrier. Moreover, in these other cases V_{ext} is a function of r. Then, among the statements above, only (a) is correct, i.e., for large kinetic energy the sticking probability is still nearly unity. However, if W is small (<1 Mev, say), it is necessary to take into account the variation of V_{ext} over the diffuse nuclear boundary. This will be done in §8 with the result that the formation probability remains of the order unity even for W small, i.e. even when the energy of the incident particle is only just sufficient to go over the top of the barrier.

In the next section we shall discuss the connection between our ζ and the observable cross sections as well as the level widths occurring in dispersion theory. In the following sections we shall consider the cases of slow neutrons (§6), of particles going through a potential barrier (§7), of particles just able to go over the top of a barrier (§8) and of very fast particles (§9). Finally, we shall investigate the influence of a nonvanishing nuclear potential V_n (§10).

§5. Cross Section, Level Width and Formation Probability

From the formulas given in the preceding section, we can compute the formation proba-

bility ζ for a given energy E of the incident particle as a function of the orbital momentum l. The probability ζ_l is defined as the fraction of the incident particles of the given angular momentum which are absorbed by the nucleus. The total number of incident particles of orbital momentum l per second is $(2l+1)\pi\lambda\!\!\!^{-2}$, if the total incident current is unity and $\lambda\!\!\!^{-}$ is the wavelength of the particle at large distance from the nucleus. The total cross section for all possible processes is therefore

$$\sigma_t=\sum_{l=0}^{\infty}\sigma_l=\pi\lambda\!\!\!^{-2}\sum_l(2l+1)\zeta_l. \tag{20}$$

If the fundamental assumption of the theory of the compound nucleus is correct, i.e., if the disintegration of the compound nucleus is independent of the way of its formation, the partial cross section for a certain process A is obtained by the well-known formula

$$\sigma_A=\sigma_t\gamma_A/\gamma, \tag{20a}$$

where γ_A is the partial width for the given process and γ the total width. Both γ_A and γ represent averages over all compound levels near the energy of the incident particle.

Equation (20) does not include the whole elastic scattering, but only the part due to formation of the compound nucleus and reemission of the incident particle without phase relations [cf. Eqs. (20), (20a)]. This part which corresponds to the resonance scattering in the region of separated levels, is small if many other processes (inelastic scattering or disintegrations) can occur. The remaining part of the elastic scattering follows from the asymptotic behavior of the radial wave function u. For a free particle, the radial wave of orbital momentum l behaves asymptotically as

$$e^{-ikr}+(-1)^{l+1}e^{ikr}. \tag{21a}$$

If the actual wave function behaves as

$$e^{-ikr}+(-1)^{l+1}\beta_l e^{ikr}, \tag{21b}$$

the scattered amplitude will be

$$F(\vartheta)=-\tfrac{1}{2}i\lambda\!\!\!^{-}\sum_l(2l+1)(\beta_l-1)P_l(\cos\vartheta). \tag{21}$$

The total elastic cross section is then

$$\sigma_{el}=2\pi\int|F|^2\sin\vartheta d\vartheta =\pi\lambda\!\!\!^{-2}\sum_l(2l+1)|\beta_l-1|^2. \tag{22}$$

From Eq. (17) we have, taking into account the phase shift outside the nucleus[15a] due to V_{ext}

$$\beta=\exp\{\pi il-2l^{\frac{1}{2}}(l+1)^{\frac{1}{2}}\arctan[l^{\frac{1}{2}}(l+1)^{\frac{1}{2}}/kR] -2ikR+2i\eta-\pi kb\} \tag{22a}$$

β is related to the formation probability by

$$\zeta_l=1-|\beta_l|^2. \tag{22b}$$

The cross section for formation of the compound nucleus by incident particles of orbital momentum l is $(2l+1)\pi\lambda\!\!\!^{-2}\zeta_l$. Adding to this the contribution l to the elastic cross section (22), we find for the l part of the total cross section (elastic plus inelastic)

$$s_l=(2l+1)\pi\lambda\!\!\!^{-2}\cdot 2(1-\mathrm{Re}\,\beta_l), \tag{23}$$

Re denoting the real part. Comparing this with (21), we find

$$s_l=4\pi\lambda\!\!\!^{-}\,\mathrm{Im}\,F_l, \tag{24}$$

where F_l is the factor of P_l in (21) and Im denotes the imaginary part. The relation (24) between the total cross section and the imaginary part of the elastically scattered amplitude was found by Bohr, Peierls and Placzek[5] and will be referred to below as the BPP theorem.

According to Eq. (22a), β is very small as long as W is positive and greater than about 1 Mev. For fast neutrons this is the case if $l<R/\lambda\!\!\!^{-}$. On the other hand, we shall show in §7 that β is nearly unity for W negative and $|W|$ greater than about 1 Mev, i.e. for $l>R/\lambda\!\!\!^{-}$. The intermediate region, -1 Mev$<W<1$ Mev, is not very important for high E. Therefore [cf. Eq. (20)] the total cross section for all inelastic processes is about πR^2 and the cross section for elastic nonresonance scattering [cf. Eq. (22)] is also equal to πR^2. This latter scattering represents the diffraction of the geometrical shadow of the nucleus, as has been pointed out by

[15a] This expression was calculated by S. P. Frankel of the University of Rochester to whom I am indebted for its communication.

Bohr, Peierls and Placzek;[5] the "shadow scattering" goes mostly in the forward direction.[5, 11, 11a]

In the case of *separated compound levels*, it is important to know the average width of the levels. This quantity must also be known in the case of overlapping levels when we want to calculate the partial cross sections [cf. Eq. (20a)]. From statistical arguments (cf. reference 8), we can deduce the relation

$$\langle\Gamma_{Al}{}^{J}\rangle_{\text{Av}}=D_J\frac{(2i+1)(2s+1)}{2J+1}\frac{\sigma_{lJ}}{2\pi^2\lambda^2}. \tag{25}$$

Here $\Gamma_{Al}{}^{J}$ represents the partial width for disintegration of a compound level of angular momentum J into a particle P of orbital momentum l and a residual nucleus in the quantum state A; i and s are the spins of residual nucleus and particle P, respectively; σ_{lJ} is the cross section for formation of the compound nucleus in the inverse process, i.e. of a compound nucleus of angular momentum J by an incident particle of orbital momentum l; λ is the wave-length of the particle emitted, and D_J is the average spacing between the levels of angular momentum J of the compound nucleus.

We may write

$$\sigma_{lJ}=\sigma_l\,\alpha_{lJ}, \tag{25a}$$

where σ_l is the total cross section for formation of the compound nucleus by particles of orbital momentum l, and α_{lJ} the probability that the nucleus so formed has angular momentum J. If the nuclear reaction is statistical with respect to angular momentum, i.e. if ζ does not depend on the total angular momentum J (which must, e.g., be true if $\zeta=1$), we have

$$\alpha_{lJ}=g_{li_s,J}(2J+1)/(2l+1)(2i+1)(2s+1), \tag{25b}$$

where $g_{li_s,J}$ is an integer giving the number of ways in which the resultant J can be obtained by compounding l, i and s vectorially. If one of the three numbers l, i, s is zero, g is unity for all J's which can be obtained at all. If $s=\frac{1}{2}$, as for proton and neutron, g is either one or two, *viz.*

$$\begin{aligned} &g_{li_s,J}=1 \quad \text{for} \quad J=l+i+\tfrac{1}{2} \quad \text{and} \quad |l-i|-\tfrac{1}{2} \\ &g_{li_s,J}=2 \quad \text{for} \quad |l-i|+\tfrac{1}{2}\leqslant J\leqslant l+i-\tfrac{1}{2}. \end{aligned} \tag{25c}$$

For $l=i$, the level $J=|l-i|-\frac{1}{2}$ does, of course, not exist.

Inserting (25a), (25b) and (20) into (25), we find

$$\langle\Gamma_{Al}{}^{J}\rangle_{\text{Av}}=g_{li_s,J}D_J\,\zeta/2\pi. \tag{26}$$

This equation contains the well-known result that the partial width of a compound level for a given mode of disintegration is of the order of the spacing between levels if the formation probability ζ is of the order unity.

§6. Slow Neutrons. Relation to the Dispersion Theory

A. Capture

It has been shown in §4 that the formation probability of the compound nucleus for slow neutrons of zero orbital momentum is [cf. Eq. (18a)]

$$\zeta=2\pi b/\lambda. \tag{27}$$

The total cross section exclusive of potential scattering is then [cf. Eq. (20), $l=0$]

$$\sigma_l=2\pi^2\lambda b. \tag{27a}$$

This formula contains the $1/v$ law. It is, of course, only valid when the energy levels of the compound nucleus overlap. The average neutron width $\langle\Gamma_N\rangle_{\text{Av}}$ of the energy levels can be obtained from (26). Since $l=0$, we have $g_{li_s,J}=1$ and therefore

$$\langle\Gamma_N\rangle_{\text{Av}}=Db/\lambda. \tag{28}$$

With $b=0.86\times10^{-13}$ cm, this gives

$$\langle\Gamma_N\rangle_{\text{Av}}=1.9\times10^{-4}DE^{\frac{1}{2}}, \tag{28a}$$

if E is the neutron energy in ev. If we assume for D a value of 10 ev which seems of the right order for medium heavy nuclei ($A\approx100$), $\langle\Gamma_N\rangle_{\text{Av}}E^{-\frac{1}{2}}$ comes out about 2×10^{-3} volt$^{\frac{1}{2}}$ which is in fair agreement with the observed values (reference 13, p. 150). Values of Γ_N for individual levels will, of course, differ considerably from the average.

Our result for the neutron width is rather larger than was believed in the beginnings of slow neutron physics. On the other hand, the radiation width Γ_γ of the compound levels seems to be rather smaller than was believed originally. In practically all cases of medium heavy nuclei,

Γ_γ seems to be smaller than 0.1 ev, and in some cases considerably so. If $\langle\Gamma_\gamma\rangle_{Av}=0.05$ ev and $D=10$ ev, the average neutron width (28a) becomes greater than the average radiation width already for neutron energies as low as 1000 ev. For lighter nuclei, let us say $A\approx 50$, the spacing D will be considerably greater while $\langle\Gamma_\gamma\rangle_{Av}$ will be only slightly larger than for $A\approx 100$; then the neutron width will, in general, be larger than the radiation width even for energies of the order of D.

The cross section for neutron capture in the region of separated levels can be calculated from the dispersion formula of Peierls and Kapur.[4] Our theory adds to dispersion theory only the formula (28) for the average neutron width.

We can make more direct predictions from our theory for neutrons of medium energy, such as the photoneutrons produced in deuterium by the γ-rays from ThC'' (neutron energy ~ 225 kev). In this energy region we can use the formulae for the cross section averaged over the resonances. For the radiative capture of the neutrons the averaged cross section is

$$\sigma_c=2\pi^2\lambda^2\langle\Gamma_N\Gamma_\gamma/D\Gamma\rangle_{Av}, \tag{29}$$

where Γ_N is the partial width for disintegration of the compound nucleus into a neutron and a residual nucleus in the ground state, Γ_γ the radiation width and Γ the total width, all quantities being averaged over the compound levels. As we have pointed out above, the neutron width can be expected to be considerably larger than the radiation width for a neutron energy of 225 kev. Therefore the total width Γ is determined by the neutron width, and we may write

$$\Gamma/\Gamma_N=N(E)=\sum_n(E-W_n)^{\frac{1}{2}}/E^{\frac{1}{2}}. \tag{29a}$$

Here n labels the energy levels of the initial (not the compound) nucleus whose excitation energy W_n is less than the kinetic energy E of the incident neutron. The partial width for disintegration into a neutron and a residual nucleus in state n, has been assumed to be simply proportional to the square root of the kinetic energy of the outgoing neutron, in accord with (26), (27) and remembering that $g_{li_n,J}=1$ because $l=0$.

Inserting (29a) in (29), we obtain

$$\sigma_c=\frac{4.1\times 10^{-24}}{EN(E)}\frac{\Gamma_\gamma}{D}\ \text{cm}^2, \tag{29b}$$

where E is measured in Mev. $N(E)$ will in general be of the order unity because E is rather low. Taking $E=0.22_5$ Mev, $\Gamma_\gamma=0.05$ ev, $D=10$ ev and $N(E)=1$, we get $\sigma_c=9\times 10^{-26}$ cm^2. The cross sections observed by v. Halban and Kowarski[16] vary considerably from one element to another, the largest ones being of the order of magnitude predicted by our theory, *viz.* about $2-5\times 10^{-26}$ cm^2. The cross sections of the lighter elements ($A\approx 60$) are consistently smaller, *viz.* between 3×10^{-28} and 10^{-27} cm^2, which is readily explained by a greater spacing D between the energy levels. However, some of the nuclei of $A\approx 100-150$ also have small cross sections of the order of 10^{-27} cm^2; whether this is due to an unusually large D or to a large $N(E)$, cannot be decided at present.

B. Scattering

Multiplying (17) by e^{-ikR}, we obtain

$$ue^{-ikR}=e^{-ikr}-e^{ikr}e^{-2ikR-\pi kb+2i\eta}. \tag{30}$$

The amplitude of the elastically scattered wave is then (cf. (21); in our case, $k=1/\lambda$)

$$F=(e^{-2ikR-\pi kb+2i\eta}-1)/2ik. \tag{30a}$$

η is [cf. Eq. (17a)] the complex phase of $(2ikb)!$; since kb is supposed to be small, we may write

$$\eta=2kb\psi(0)=-2kbC, \tag{30b}$$

where ψ is the logarithmic derivative of the factorial function and $C=0.577\cdots$ Euler's constant. Then, again remembering that k is small, (30a) becomes

$$F=-(R+2Cb)+\tfrac{1}{2}i\pi b. \tag{31}$$

The scattered amplitude (31) contains a real and an imaginary part. The former is, in sign and magnitude, the same as for a hard sphere of radius

$$R'=R+2Cb. \tag{31a}$$

[16] H. v. Halban and L. Kowarski, Nature **142**, 392 (1938).

This part is usually called potential scattering. It was originally explained in a very artificial way,[2] using a repulsive potential. Later, it was shown to come out naturally from the theory of Peierls and Kapur[4] but the result was somewhat uncertain because of the contribution of the distant levels (cf. 32c). It is now seen to result equally naturally from the assumption of a strong absorption in the nucleus. Our derivation has the advantage of giving a more definite physical meaning to the nuclear radius. The effective radius, R', is slightly larger than the R defined above. We may define R' directly as that radius for which the absorption potential is

$$\sigma(R')=(\hbar^2/2mb^2)e^{-2G}, \tag{31b}$$

which, for $b=0.86\times10^{-13}$ cm, is about 9 Mev. Mathematically, R' can be considered as the point where the WKB treatment of σ breaks down. The sign of the potential scattering should be the same (negative) for all nuclei.

The scattered amplitude F (31) also has an imaginary part. This is connected with the probability of neutron capture because it derives from the factor $e^{-\pi kb}$ in (30a) whose square is equal to one minus the formation probability ζ [cf. Eqs. (17b), (18)]. The relation between the imaginary part of (31) and the cross section for formation of the compound nucleus is a special case of the theorem of Bohr, Peierls and Placzek [cf. Eq. (24)]. (In our case, the formation cross section is practically equal to the total cross section because it contains the factor $1/v$). In the region of overlapping levels where (31) is valid, its imaginary part is not very important for the total scattering, in comparison with the real part. The total elastic scattering cross section is then

$$\sigma_s=4\pi|F|^2=4\pi(R'^2+\tfrac{1}{4}\pi^2b^2). \tag{31c}$$

We shall now discuss the scattering according to the dispersion formula, and compare it with the results just obtained for overlapping levels. According to Peierls and Kapur [reference 4, Eq. (24)], the scattered amplitude is approximately[17]

$$F=\frac{1}{2ik}\left[e^{-2ikR''}\left(1-i\sum_n\frac{\gamma_{nN}}{E-E_n+\frac{1}{2}i\gamma_n}\right)-1\right], \tag{32}$$

where R'' is the nuclear radius introduced by Peierls and Kapur (their r_0), E_n is the neutron energy which gives exact resonance with the nth compound level, γ_n is the total width of the level, and γ_{nN} its partial width for disintegration into a neutron and a residual nucleus in the ground state. For small k,

$$F=-R''-\tfrac{1}{2}\lambda\sum_n\frac{\gamma_{nN}}{E-E_n+\frac{1}{2}i\gamma_n}. \tag{32a}$$

This expression can be separated into a real and an imaginary part:

$$F=F_r+iF_i, \tag{32b}$$

$$F_r=-R''-\tfrac{1}{2}\lambda\sum_n\frac{\gamma_{nN}(E-E_n)}{(E-E_n)^2+\frac{1}{4}\gamma_n^2}, \tag{32c}$$

$$F_i=\tfrac{1}{4}\lambda\sum_n\frac{\gamma_n\gamma_{n\Lambda}}{(E-E_n)^2+\frac{1}{4}\gamma_n^2}. \tag{32d}$$

The exact theory of Peierls and Kapur is still valid when the compound levels overlap. In this case, our theory using an absorption potential is also valid, and a comparison can be made. In the case of overlapping levels the theory of Peierls and Kapur simplifies to (cf. reference 5)

$$F_i=\tfrac{1}{2}\pi\lambda\langle\gamma_N\rangle_{Av}/D, \tag{33}$$

the average being taken over the compound levels near the neutron energy E. (Γ is the value of γ at the resonance energy, cf. reference 2.) With (28), this gives

$$F_i=\tfrac{1}{2}\pi b \tag{33a}$$

in exact agreement with Eq. (31). This agreement is not surprising and means only that both the dispersion theory and our theory satisfy the theorem of Bohr, Peierls and Placzek [Eq. (24)].

The sum in the real part of the Peierls-Kapur scattered amplitude consists of the contribution of the levels close to E and of the distant levels just as (32c). The former can be shown to be zero when the levels overlap.[5] The latter is almost independent of the energy and therefore

[17] Since we shall apply (32) only to the case of low energies, we have put the u_n^2/N_n of Peierls and Kapur equal to $|u_n|^2=\gamma_{nN}$.

represents simply an addition to R''. Since the Peierls-Kapur theory is a consistent treatment of the compound nucleus, the final result, i.e. R'' *plus* the contribution of the distant levels, must be independent of the (arbitrary) choice of R''. Comparing (32c) with (31), (31a) we find that for overlapping levels

$$F_r = -R'. \tag{34}$$

In other words, *the contribution of the distant levels* in (33a) is *zero when R'' is chosen equal to R'*. Thus our theory provides a natural choice for the nuclear radius in the Peierls-Kapur theory. Since the contribution of distant levels is almost independent of E, we can write in the region of separated levels

$$F_r = -R' - \tfrac{1}{2}\lambda \sum_n \frac{\gamma_{nN}(E-E_n)}{(E-E_n)^2+\tfrac{1}{4}\gamma_n{}^2}, \tag{34a}$$

where the sum is to be extended only over the levels close to E. Taking into account the conservation of angular momentum, we obtain then for the elastic scattering cross section the value given in Eq. (54) of reference 2, with R replaced by R'.

It is interesting to estimate the relative importance of resonance and potential scattering. In the region of overlapping levels we have seen that the potential scattering is considerably larger than the resonance scattering [cf. Eq. (31c)]. (The overlapping of levels will certainly occur for neutron energies above 1 Mev because then the partial neutron width is greater than the spacing; it may already occur at lower energies if there are many low excited levels of the initial nucleus.) In the region of separated levels, the average of the elastic scattering cross section over an energy interval large compared with D, is

$$\langle\sigma_{el}\rangle_{Av} = 4\pi(R'^2+\tfrac{1}{2}\pi\lambda^2\langle\Gamma_N\rangle_{Av}{}^2/\Gamma_{Av}D) \tag{35}$$

$$= 4\pi(R'^2+\tfrac{1}{2}\pi\lambda b\langle\Gamma_N\rangle_{Av}/\Gamma_{Av}). \tag{35a}$$

If the neutron energy E is high enough so that the partial neutron width is greater than the radiation width (i.e. perhaps for $E>1$ kev for medium heavy nuclei), and if, on the other hand, E is smaller than the first excited level of the initial nucleus, the total level width Γ is nearly equal to the partial neutron width Γ_N. Then

$$\langle\sigma_{el}\rangle_{Av} = 4\pi(R'^2+\tfrac{1}{2}\pi\lambda b). \tag{35b}$$

If we take $R'=6\times10^{-13}$ cm (medium heavy nuclei) and $b=0.86\times10^{-13}$ cm, the second term in (35b) (average resonance scattering) becomes greater than the first (potential scattering) for $E<30$ kev. Therefore, according to (35b), we should have an energy region (from about 1 to 30 kev) in which the average elastic scattering cross section is proportional to $1/v$.

For smaller energies (below about 1 kev) we have $\Gamma_\gamma > \Gamma_N$; therefore the elastic cross section, averaged over the neutron energy, becomes independent of E, *viz.*

$$\langle\sigma_{el}\rangle_{Av} = 4\pi(R'^2+\tfrac{1}{2}\pi b^2 D/\Gamma_\gamma). \tag{35c}$$

With our assumed values for b, D and Γ_γ, the second term gives a cross section of 30×10^{-24} cm^2. It is not surprising that the scattering cross section for thermal neutrons is considerably smaller than this amount for practically all nuclei. This is because the main contribution to the average cross section (35c) comes from the immediate neighborhood of the resonances, and the thermal region is in general "between resonances." If we estimate the scattering amplitude (32a) between resonances by assuming $E-E_n=\frac{1}{2}D$ for one level n, and no contribution from other levels, we obtain for the resonance scattering $\approx\lambda\gamma_{nN}/D=b$. Therefore "between resonances" the potential scattering will probably be the more important term, although it must be admitted that our estimate of the resonance scattering is extremely uncertain. It would be very important to investigate experimentally the relative importance of resonance and potential scattering for slow neutrons. This could be done by interference experiments because the potential scattering amplitude should have the same sign (negative) for all nuclei while the resonance scattering may be positive or negative, according to the position of the nearest levels. Moreover, the resonance scattering is partly incoherent.[18,19]

[18] G. C. Wick, Physik. Zeits. **38**, 689 (1937).
[19] O. Halpern and M. H. Johnson, Phys. Rev. **55**, 898 (1939).

§7. Potential Barrier

In this section, we shall treat the case of a particle which must penetrate a potential barrier in order to enter the nucleus. In deriving Eq. (17), it was not assumed that the kinetic energy W at the surface of the nucleus was positive. If W is negative, we may simply put

$$k=i\kappa, \tag{36}$$

where

$$\kappa^2=-2mW/\hbar^2. \tag{36a}$$

Then (17) goes over into

$$u=e^{\kappa(r-R)}-e^{-\kappa(r-R)}\frac{(-\kappa b)!}{(\kappa b)!}e^{-\pi i\kappa b}. \tag{37}$$

Since the kinetic energy $E-V_{\text{ext}}$ [cf. Eq. (4)] varies with r, this expression must be replaced by the WKB expression

$$u(r)=|\Phi|^{-\frac{1}{4}}\exp\left(\int_R^r|\Phi(\rho)|^{\frac{1}{2}}d\rho\right)-|\Phi|^{-\frac{1}{4}}e^{-\pi i\kappa b}\exp\left(-\int_R^r|\Phi(\rho)|^{\frac{1}{2}}d\rho\right)(-\kappa b)!/(\kappa b)!, \tag{37a}$$

where

$$\Phi=(2m/\hbar^2)(E-V_{\text{ext}}). \tag{37b}$$

For large r, Φ becomes positive and (37a) goes over into

$$u=2\Phi^{-\frac{1}{4}}\sin\left(\int_{r_0}^r\Phi^{\frac{1}{2}}d\rho+\tfrac{1}{4}\pi\right)\exp\left(\int_R^{r_0}|\Phi(\rho)|^{\frac{1}{2}}d\rho\right)$$

$$-\Phi^{-\frac{1}{4}}\cos\left(\int_{r_0}^r\Phi^{\frac{1}{2}}d\rho+\tfrac{1}{4}\pi\right)\exp\left(-\int_R^{r_0}|\Phi(\rho)|^{\frac{1}{2}}d\rho\right)e^{-\pi i\kappa b}(-\kappa b)!/(\kappa b)!, \tag{37c}$$

where r_0 is the point at which Φ is zero and ρ an integration variable. The function (37c) can be represented as an incident plus an outgoing wave. The reflection coefficient can be calculated in the usual way as the absolute square of the ratio of the amplitudes of the two waves. The formation probability then is one minus the reflection coefficient and comes out to be

$$\zeta=2\sin\pi\kappa b\;e^{-2G}(-\kappa b)!/(\kappa b)!+0(e^{-4G}) \tag{38}$$

with

$$G=\int_R^{r_0}|\Phi(r)|^{\frac{1}{2}}dr, \tag{38a}$$

the Gamow integral. Using the well-known relation between $(-n)!$ and $n!$ (cf. reference 15, p. 89), we find

$$\zeta=2\pi\kappa be^{-2G}/(\kappa b)!^2 \tag{38b}$$

The formation probability ζ, as here defined, includes the penetrability of the potential barrier (factor e^{-2G}) and differs in this respect from the sticking probability ξ used commonly in the literature (cf. §3B). For small κb, the ζ from (38b) is very nearly $2\pi\kappa be^{-2G}$, i.e. it contains, besides the penetrability factor e^{-2G}, another factor which is analogous to the formation probability for small positive W, *viz.* $2\pi kb$. If we neglect the variation of V_{ext} with r, the formation probability drops from 1 at high energies to 0 at $W=0$, then rises again for negative W as $|W|^{\frac{1}{2}}$, and finally decreases again because of the factor e^{-2G}.

Our formula for ζ is in agreement with the result of Peierls and Kapur,[4] which has also been used by Weisskopf and Ewing,[8] namely that the probability of formation of the compound nucleus is inversely proportional to the square of the irregular solution of the wave equation in the Coulomb field. For $r<r_0$ this

solution behaves as

$$|\Phi|^{-\frac{1}{2}}(r)\exp\left[\int_r^{r_0}|\phi(\rho)|^{\frac{1}{2}}d\rho\right]$$

which makes the formation probability proportional to $|\Phi(R)|^{\frac{1}{2}}e^{-2G}=\kappa e^{-2G}$, in agreement with our result. Previously, it had been supposed that the formation probability was directly proportional to the square of the regular solution, i.e. $\sim(1/\kappa)e^{-2G}$, or else that it contained only[20] the exponential e^{-2G} without any power of κ. However, it must be remembered that the variation of V_{ext} with r invalidates our Eq. (38b) for small κ for which the factor κ would be most important. We shall show in the next section that, because of the variation of V_{ext}, ζ does not actually go to zero for $W=0$ and then rise again to a secondary maximum for negative W, but that it is of the order unity for $W=0$ and decreases monotonically with decreasing W. Therefore the simple exponential e^{-2G} is probably a better approximation to ζ than the complete formula (38b), at least as long as the factor $2\pi\kappa b$ is smaller than unity.

For very high barrier (W large and negative), (38b) decreases faster than the penetrability e^{-2G}, because of the denominator $(\kappa b)!^2$. This result is spurious because it depends essentially on the assumption that V_{ext} is constant everywhere. A more appropriate treatment of the case of large negative W can be made on the basis of the WKB which in this case is a very good approximation.

The WKB gives for all $r<r_0$:

$$u=c(V-E-i\sigma)^{-\frac{1}{4}}\exp\left\{(2m)^{\frac{1}{2}}\hbar^{-1}\int_{r_0}^{r}(V(\rho)-E-i\sigma(\rho))^{\frac{1}{2}}d\rho\right\}, \tag{39}$$

if we write for brevity V instead of V_{ext}. Then the wave function at large distances ($r\gg r_0$, $V\ll E$) is

$$u=2cE^{-\frac{1}{4}}\cos(2mE/\hbar^2)^{\frac{1}{2}}r. \tag{39a}$$

It will be most convenient to normalize to unit incident current, which means $c=(m/2)^{\frac{1}{4}}$. The total absorption per second can in our case be most easily calculated from the definition (1) of σ; we have

$$\zeta=(2/\hbar)\int_0^\infty|u(r)|^2\sigma(r)dr. \tag{40}$$

If we insert (13) for σ and (39) for u, and neglect σ^2 compared with $(V-E)^2$ in u, we obtain

$$\zeta=\frac{\hbar}{(2m)^{\frac{1}{2}}(V-E)^{\frac{1}{2}}b^2}\int_0^\infty dr\exp\left\{-\frac{r-R}{b}+2\frac{(2m)^{\frac{1}{2}}}{\hbar}\int_{r_0}^{r}(V-E)^{\frac{1}{2}}d\rho\right\}. \tag{40a}$$

The exponential in (40a) has a maximum for $r=r_1$ where r_1 is defined by

$$V(r_1)-E=\hbar^2/8mb^2. \tag{40b}$$

In order to justify our neglect of σ compared with $V-E$ in the integrand of (40a), we must require that r_1 be appreciably greater than R, in other words that $W(R)>\hbar^2/8mb^2$. For our value $b=0.86\times10^{-13}$ cm, we have $\hbar^2/8mb^2=7$ Mev for a proton or neutron. We note that the condition $W>\hbar^2/8mb^2$ is equivalent to $\kappa b>\frac{1}{2}$; for smaller values of κb, (38b) can be used, and the $(\kappa b)!$ in that formula can be replaced by unity.

If $W>\hbar^2/8mb^2$, there will be a certain $r_1>R$ for which (40b) is fulfilled. If we expand the exponent in (40a) in powers of $r-r_1$ up to the quadratic terms and integrate, we obtain, remembering (40b)

$$\zeta=\left(\frac{2\pi\hbar^2}{mb^3}\right)^{\frac{1}{2}}\left(\frac{dV}{dr}\right)_{r_1}^{-\frac{1}{2}}\exp\left\{-2G+2\frac{(2m)^{\frac{1}{2}}}{\hbar}\int_R^{r_1}(V-E)^{\frac{1}{2}}dr-\frac{r_1-R}{b}\right\}. \tag{41}$$

[20] E. J. Konopinski and H. A. Bethe, Phys. Rev. **54**, 130 (1938).

It can easily be seen that the exponential in (41) is greater than e^{-2G}. The difference arises from the fact that the main absorption occurs not at the boundary of the nucleus but farther out (at r_1) because the square of the wave function, $|u|^2$, increases faster from R to r_1 than σ decreases, a behavior similar to the Oppenheimer-Phillips process. The effect depends, of course, essentially on the exponential decrease of σ outside the nucleus; if σ decreased more strongly, e.g., as $e^{-(r-R)^2}$, the main absorption would always be in the neighborhood of the nucleus.

Because of its dependence on the model, the deviation of ζ from the simple exponential e^{-2G} should not be taken too seriously; but apart from this, the deviation is never appreciable compared with e^{-2G} itself. If we assume $V-E$ to vary as $1/r$ the additional terms in the exponent in (41) have the value

$$\gamma_1=(R/b)(\nu^{\frac{1}{2}}-1)^2, \tag{41a}$$

where

$$\nu=W/(\hbar^2/8mb^2). \tag{41b}$$

If $V-E$ varies as $1/r^2$, we have instead of (41a)

$$\gamma_2=(R/b)(\tfrac{1}{2}\nu^{\frac{1}{2}}\log\nu-\nu^{\frac{1}{2}}+1). \tag{41c}$$

Both (41a) and (41c) are relatively small. E.g., for $\nu=2$, i.e. $W=14$ Mev, we have $\gamma_1=0.17R/b$ and $\gamma_2=0.077R/b$. Since R/b is only 10 for the heaviest nuclei, the exponential differs from e^{-2G} only by a factor $e^{1.7}$ or $e^{0.77}$, respectively, which is not important compared with the large factor e^{-2G}. It is very unlikely that the additional terms in the exponential in (41) can ever be tested experimentally. Thus far, no experiments at all have been made in which the energy of the incident particle was more than $\hbar^2/8mb^2\approx 7$ Mev below the top of the potential barrier.

Summarizing, we find that (38b) is valid for moderate negative energies, perhaps from $\kappa b=1/2\pi$ to $\kappa b=\frac{1}{2}$ or $W=-0.7$ to -7 Mev. The value of ζ for greater $|W|$ is of no practical importance and depends on the exact behavior of σ as a function of r.

Our formulae for ζ can, of course, be used both to obtain the total cross section for all possible reactions in the case of overlapping levels, according to (20), and also the average width of resonance levels if such exist, according to (26).

The elastic scattering can be obtained from the behavior of the wave function for large r. It must be remembered, of course, that the Coulomb potential in the case of charged particles produces scattering by itself. Let δ be the phase shift due to the Coulomb potential for a given l ($\delta=0$ for neutrons), then the actual scattered amplitude for this l is

$$F_l=-\tfrac{1}{2}i\lambda\{e^{2i\delta}[1-ie^{-\pi i\kappa b}e^{-2G}(-\kappa b)!/(\kappa b)!]-1\}(2l+1)). \tag{42}$$

Terms of order e^{-4G} have been neglected. The differential scattering cross section per unit solid angle is $|\sum_l F_l P_l(\cos\vartheta)|^2$. The term proportional to e^{-2G} in (42) represents the nuclear scattering. For neutrons, this term alone exists, and we have

$$F_l=-(l+\tfrac{1}{2})\lambda e^{-2G}e^{-\pi i\kappa b}(-\kappa b)!/(\kappa b)! \tag{42a}$$

(The potential V_{ext} for neutrons is simply the centrifugal potential, $\hbar^2 l(l+1)/2mr^2$.) The expression (42a) is nearly real for small κb and corresponds then to the potential scattering found for small positive W (§6B). The real part decreases in relative importance as κb increases and reaches zero for $\kappa b=\frac{1}{2}$. Then there remains only the imaginary part which is required by the theorem of Bohr, Peierls and Placzek, and is equal to

$$F_l=\tfrac{1}{2}i(l+\tfrac{1}{2})\lambda\zeta. \tag{42b}$$

For $\kappa b>\frac{1}{2}$, (42a) is no longer valid, for the same reasons as (38). However, the WKB is then very nearly valid; and it can easily be shown that F_l is purely imaginary when terms of the order $\sigma^2/(V-E)^2$ are neglected [cf. above, Eq. (40) ff.]. Therefore (42b) remains valid; the elastic scattering at very large negative kinetic

energies, just as at large positive energies, is only the amount necessary because of the existence of inelastic processes according to the optical theorem.

§8. Spatial Variation of the External Potential

With the exception of the case of neutrons of zero orbital momentum, the extranuclear potential V_{ext} changes (decreases) with r. Therefore, if W is very small, $V_{ext}-E$ will change by its own amount and more over a very short distance. In this case, it is not justified to consider $V_{ext}-E$ as constant over the range of action of the nuclear absorption coefficient σ, as has been done in §4–7. It will, however, be sufficient to consider $V_{ext}-E$ as changing linearly with r. We shall put

$$(2m/\hbar^2)(E-V_{ext})=k^2+A(r-R'), \quad (43)$$

where A is a positive constant measuring the force on the particle,

$$A=-(2m/\hbar^2)(dV/dr)_{R'}, \quad (43a)$$

and R' is the radius of the hard sphere which is equivalent to the nucleus with regard to the potential scattering of slow neutrons (§6B). It is more convenient to use R' than R for our purpose. $(\hbar^2/2m)k^2$ is the kinetic energy at $r=R'$.

At large distances from the nucleus, σ is negligible and we have

$$d^2u/dr^2+[k^2+A(r-R')]u=0. \quad (44)$$

Here we introduce the abbreviation

$$x=r-R'+k^2/A. \quad (44a)$$

The solution of (44) is well known and has been investigated in detail by Kramers[21] in connection with the WKB method. The regular solution which vanishes for large negative x and is identical with the solution investigated by Kramers, is given by

$$v=\tfrac{1}{3}(\pi x)^{\frac{1}{2}}\{J_{1/3}(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{3}{2}})+J_{-1/3}(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{3}{2}})\}, \quad (45)$$

where J is the Bessel function. The function v is so normalized as to go over, for large x, into the WKB solution

$$v\rightarrow(Ax)^{-\frac{1}{4}}\cos(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{3}{2}}-\tfrac{1}{4}\pi). \quad (45a)$$

For negative x, the analytical continuation of (45) is

$$v=\tfrac{1}{2}(\pi|x|/3)^{\frac{1}{2}}e^{2\pi i/3}H_{1/3}^{(1)}(\tfrac{2}{3}A^{\frac{1}{2}}i|x|^{\frac{3}{2}}). \quad (45b)$$

The irregular solution of (44) which is shifted in phase by $\pi/2$ against v, is the function

$$w=3^{-\frac{1}{2}}(\pi x)^{\frac{1}{2}}\{J_{-1/3}(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{3}{2}})-J_{1/3}(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{3}{2}})\}, \quad (46)$$

whose asymptotic behavior for large x is

$$w\rightarrow(Ax)^{-\frac{1}{4}}\cos(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{3}{2}}+\tfrac{1}{4}\pi). \quad (46a)$$

The general solution may be written

$$u=v+\alpha w. \quad (47)$$

Then the ratio of the amplitude of the outgoing to that of the incident wave is

$$\beta=(1+i\alpha)/(1-i\alpha), \quad (47a)$$

the reflection coefficient is $R=|\beta|^2$ and the scattered amplitude is proportional to $\beta-1$. Therefore we need only determine α. Inserting (45), (46), and omitting a constant factor, we may write (47) in the form

$$u\sim x^{\frac{1}{2}}[(1+\alpha\sqrt{3})J_{-1/3}(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{3}{2}})+(1-\alpha\sqrt{3})J_{1/3}(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{3}{2}})]. \quad (48)$$

We shall be interested in the behavior of u for $r\approx R'$, and small k^2, i.e., for small x [cf. Eq. (44a)].

[21] H. A. Kramers, Zeits. f. Physik **39**, 828 (1926).

In this case we have

$$x^{\frac{1}{2}}J_{-1/3}(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{1}{2}})=3^{\frac{1}{2}}A^{-\frac{1}{6}}(-\tfrac{1}{3})!^{-1}(1-\tfrac{1}{6}Ax^3+\cdots), \tag{48a}$$

$$x^{\frac{1}{2}}J_{1/3}(\tfrac{2}{3}A^{\frac{1}{2}}x^{\frac{1}{2}})=3^{-\frac{1}{2}}A^{\frac{1}{6}}(\tfrac{1}{3})!^{-1}x(1-\tfrac{1}{12}Ax^3+\cdots). \tag{48b}$$

Inserting in (48) and leaving out a constant factor, we find

$$u\sim 1-\tfrac{1}{6}Ax^3+\cdots+\frac{1-\alpha\sqrt{3}}{1+\alpha\sqrt{3}}\frac{3^{\frac{1}{2}}}{2}\frac{(\frac{2}{3})!}{(\frac{1}{3})!}A^{\frac{1}{2}}x(1-\tfrac{1}{12}Ax^3+\cdots). \tag{49}$$

It will be convenient to use the abbreviation

$$\kappa=2\cdot 3^{-\frac{1}{2}}\cdot(\tfrac{1}{3})!/(\tfrac{2}{3})!=1.372. \tag{49a}$$

Kramers has given a table of the regular solution v for small values of $\xi=A^{\frac{1}{3}}x$. By numerical calculation, it can be shown that this solution differs by only $4\frac{1}{2}$ percent from the WKB value at $\xi=+1$, and by 8 percent at $\xi=-1$. It will therefore be necessary to use the more complicated theory of this section only if $|\xi|<1$ at the boundary of the nucleus ($r=R'$). This condition is equivalent to

$$|k^2|<A^{\frac{1}{3}} \tag{50}$$

or

$$|E-V_{\text{ext}}(R')|<(\hbar^2/2m)^{\frac{1}{3}}(dV/dr)^{\frac{2}{3}}. \tag{50a}$$

For $|\xi|<1$, the expressions given in (48a, b) for the J's are accurate to $\frac{1}{2}$ and $\frac{1}{5}$ percent, respectively.

Our problem is now to solve the Schrödinger equation including σ, i.e.

$$\frac{d^2u}{dr^2}+(Ax+\frac{i}{b^2}e^{-(r-R)/b})u=0. \tag{51}$$

This can, of course, not be done exactly. However, we can use the fact that k^2 is small to get a good approximation. In the region where σ is important, Ax will be relatively small and it will be sufficient to take it into account in first approximation. The solution obtained in this way will, for large r, go over automatically into the solution of the equation without σ, neglecting terms in A^2 in the latter. As pointed out in the last paragraph, this is justified for all cases in which the variation of V_{ext} with r is at all important.

Equation (51) with $A=0$ has the solution (cf. §4)

$$u_0=2H_0^{(1)}(2i^{\frac{1}{2}}e^{-(r-R)/2b}), \tag{52}$$

the factor 2 being introduced for convenience in the following. For large r, (52) behaves as

$$u_0\rightarrow-(2i/\pi)(r-R')/b. \tag{52a}$$

With the usual method, we obtain between u and u_0 the relation

$$u_0du/dr-udu_0/dr=\int_{-\infty}^{r}[A(\rho-R')+k^2]u(\rho)u_0(\rho)d\rho. \tag{53}$$

Here it has been assumed that u as well as u_0 decreases towards small r inside the nucleus. In first approximation, we may put $u=u_0$ in the integral. Then the integration is elementary for values of r large enough so that (52a) is valid. Only the integration constant cannot be determined in an elementary way; it must be a linear function of A and k^2. The coefficient of k^2 can be determined by analytical integration, that of A was found by numerical integration. Neglecting terms of order $e^{-(r-R')/b}$ and using the abbreviation

$$y=(r-R')/b, \tag{53a}$$

we obtain the result:

$$u=1-2iy/\pi+k^2b^2(\lambda y-\tfrac{1}{2}y^2+(i/3\pi)y^3)+Ab^3(\mu y-\tfrac{1}{6}y^3+(i/6\pi)y^4) \tag{54}$$

with

$$\lambda=\frac{32}{3\pi^2}\left(1+\frac{1}{2^3}+\frac{1}{3^3}+\cdots\right)-\frac{\pi}{6}i=1.298-0.523i, \tag{54a}$$

$$\mu=1.339-2.034i. \tag{54b}$$

We may now compare (49) with (54). Remembering that [cf. Eqs. (44a), (53a)]

$$x=by+k^2/A \tag{55}$$

and introducing the abbreviations

$$a=A^{\frac{1}{3}}b, \tag{55a}$$

$$v=k^2/A^{\frac{1}{3}} \tag{55b}$$

we obtain

$$\frac{1-\alpha\sqrt{3}}{1+\alpha\sqrt{3}}=\kappa\frac{-2i/\pi a+\mu a^2+\lambda av+\frac{1}{2}v^2+iv^3/3\pi a}{1+2iv/\pi a-\mu a^2v-\lambda av^2-\frac{1}{3}v^3-iv^4/6\pi a}. \tag{56}$$

Let us denote numerator and denominator of the right-hand side by ν and ρ, respectively; then the reflected amplitude β defined in (47a) is

$$\beta=\frac{\rho(\sqrt{3}+i)+\nu(\sqrt{3}-i)}{\rho(\sqrt{3}-i)+\nu(\sqrt{3}+i)}. \tag{56a}$$

Of the two constants v and a occurring in (56), v lies between the limits $+1$ and -1, because v is simply the value of $A^{\frac{1}{3}}x$ for $r=R'$, and because the WKB is valid when the absolute value of $A^{\frac{1}{3}}x$ is greater than 1 [cf. Eq. (50)]. The constant a is always positive and given by [cf. Eqs. (43a), (55a)]

$$a^3=2mb^3\hbar^{-2}|dV/dr|_{R'}. \tag{57}$$

We have

$$V(R')=Ze^2/R'+\hbar^2l(l+1)/2mR'^2, \tag{57a}$$

$$V/R'\leqslant|dV/dr|\leqslant 2V/R'. \tag{57b}$$

Since we are interested in the case of small kinetic energy $V-E$, we may put

$$|dV/dr|=\eta E/R', \tag{57c}$$

where $\eta=2$ for neutrons, $\eta=1$ for charged particles with $l=0$, and η between 1 and 2 for charged particles with $l\neq 0$. Therefore (57) becomes

$$a=(b/\lambda)^{\frac{1}{3}}(\eta b/R')^{\frac{1}{3}}. \tag{58}$$

For atomic weights between 10 and 240, the last factor is between 0.8 and 0.45. The first factor, for $b=0.86\times10^{-13}$, is equal to $(E/28\text{ Mev})^{\frac{1}{3}}$, i.e., 0.7 for 10 Mev and 0.33 for 1 Mev. Therefore a is in general less than one, and may vary between about 0.15 and 0.5.

For $v=0$, i.e. when the energy of the incident particle is exactly equal to the height of the potential barrier, the numerator and denominator of (56) reduce to

$$\nu=-2i\kappa/\pi a+\kappa\mu a^2;\quad \rho=1. \tag{51}$$

In Table I we give the value of β and of the formation probability, $\zeta=1-|\beta|^2$, for a few values of a in the important region. It is seen that ζ is of the order unity and changes only slightly with a, in the expected direction, i.e., increasing with increasing a. This shows that, due to the variation of V with r, the point $v=0$ ceases to be a singular point and is characterized by a fairly large formation probability, as has been anticipated in §7. From the value of β, the elastic scattering can be found [cf. Eq. (22)].

For $v\neq 0$, Eq. (56) is more complicated. However, it can easily be seen that $|\beta|$ decreases and

TABLE I. *Changing potential, W=0.*

a	β	ζ
small	$\frac{1}{2}-\frac{1}{2}i\sqrt{3}-1.146a(\sqrt{3}-i)$	$3.97a$
0.2	$0.369-0.570\,i$	0.539
0.3	$0.350-0.467\,i$	0.659
0.4	$0.356-0.404\,i$	0.710
0.5	$0.382-0.369\,i$	0.718

therefore ζ increases with increasing v, as it should be. Computations for $v\neq 0$ are in progress at the University of Rochester.

§9. Very High Energy

For very high energy, the formalism of the compound nucleus breaks down. Instead, the Born approximation is applicable. Because of its simplicity, this method permits a comparison between our theory and general nuclear theory. Neglecting the correlations between the various particles in the nucleus, the total cross section for all processes is from general nuclear theory:

$$\sigma_t=(A/2\pi\hbar^2v^2)\times\int_0^\infty qdq\left|\int V_n(\mathbf{r})\exp\left[i\mathbf{q}\cdot\mathbf{r}\right]d\tau\right|^2, \quad (60)$$

where $V_n(\mathbf{r})$ is the nuclear interaction potential as a function of the distance r between the interacting particles, $\hbar\mathbf{q}$ is the momentum change and v the initial velocity of the incident particle and A is the number of particles in the nucleus.

In (60), we have assumed that the interaction of very fast nuclear particles can still be described by a potential. In the following, we shall even assume that this potential V_n is the same as for the usual energies of a few Mev. There is no justification for such an assumption. However, we are not interested in the actual behavior of high energy nuclear particles but rather in the mathematical problem of the behavior of fast particles which have the interaction V_n. From the solution of this problem which can be found because of its mathematical simplicity, we can draw conclusions about the problem which forms the subject of this paper, *viz.* the behavior of particles of moderate energy having an interaction V_n with the nucleus.

The integral in (60) reduces to a quantity closely related to the binding energy of the deuteron, e.g., for an exponential potential

$$V_n(r)=Be^{-r/b}. \quad (60a)$$

(60) reduces to

$$\sigma_t=(16\pi/3)A(Bb^2/\hbar v)^2. \quad (60b)$$

According to the theory of the deuteron, with $b=0.86\times10^{-13}$ cm,

$$Bb^2=2.23\hbar^2/m. \quad (60c)$$

Considering that for the singlet potential Bb^2 is only about $1.31\hbar^2/m$, we obtain

$$\sigma_t=4\pi\lambda^2A\cdot 5.5, \quad (61)$$

where λ is the wave-length of the incident particle. This approximation is applicable when $\sigma_t\ll\pi R^2$. Putting (cf. reference 8)

$$R=r_0A^{\frac{1}{3}},\quad r_0\approx 1.3_5\cdot10^{-13}\text{ cm}, \quad (61a)$$

we obtain for the critical wave-length, at which $\sigma_t=\pi R^2$:

$$\lambda_c=r_0/(2\cdot5.5^{\frac{1}{2}}A^{\frac{1}{3}})=2.8_5\cdot10^{-14}A^{-\frac{1}{3}}\text{ cm}. \quad (61b)$$

For $A=100$, this is 1.3×10^{-14} cm and corresponds to a neutron or proton of 1200 Mev energy. The Born approximation is restricted to energies higher than this.

From our theory of the absorption potential, [cf. Eq. (1)] we have in the case of the Born approximation

$$\sigma_t=\frac{2\sigma_0}{\hbar v}\cdot\frac{4\pi}{3}R^3, \quad (62)$$

where σ_0 is the absorption potential inside the nucleus. Comparing (62) with (61), (61a), we find that in the region of the Born approximation we must have

$$\sigma_0=\frac{3}{2}\frac{\hbar^2}{mr_0^3}\cdot5.5\lambda\approx140\lambda\text{ Mev}, \quad (62a)$$

if λ is measured in 10^{-13} cm. This shows (1) that σ_0 is proportional to $1/v$ and thus changes only slowly with the energy of the incident particle, (2) that σ_0 depends on $|V_n|^2$ [cf. Eq. (60)] rather than on the average of V_n, (3) that σ_0 is very large. Point (1) has been assumed when σ was introduced in §2 and is essential in order to give validity to the dependence of the formation probability ζ on the energy as derived in this paper. Point (2) is important in order to

justify the neglect of the average nuclear potential V_n compared to the absorption potential σ (cf. the beginning of §4); the attractive and repulsive interactions existing between particles of different relative spin and charge will average out in V_n but will all give a positive contribution to σ. Finally (3) the magnitude of σ_0 may not be quite as large as would follow from (62a) for moderate energies ($\lambda\!\!\!^{-}=1-5\times10^{-13}$ cm, $E=20-1$ Mev) because (62a) has only been proved for $\lambda\!\!\!^{-}<\lambda\!\!\!^{-}_c$ [cf. Eq. (61b)]. For $\lambda\!\!\!^{-}_c\approx1.3\times10^{-14}$ cm, we have $\sigma_0=18$ Mev, and it seems safe to assume that σ_0 is greater than this value for moderate energies.

In our theory, it is quite easy to treat the transition from moderate to high energies. It is only necessary to consider the finite size of the nucleus and to make the wave function u equal to zero at $r=0$. Since we can certainly apply the WKB for these energies, we obtain

$$u(r)=\exp\left(-i(2m)^{\frac{1}{2}}\hbar^{-1}\int_0^r(E-V+i\sigma)^{\frac{1}{2}}d\rho\right)$$
$$-\exp\left(i(2m)^{\frac{1}{2}}\hbar^{-1}\int_0^r(E-V+i\sigma)^{\frac{1}{2}}d\rho\right). \quad (63)$$

Assuming σ small compared with E everywhere, and neglecting σ^2, we obtain for large r:

$$u(r)=\exp\left(-ikr-i\delta+\frac{m^{\frac{1}{2}}}{\sqrt{2}\hbar}\int_0^r\frac{\sigma(\rho)}{(E-V(\rho))^{\frac{1}{2}}}d\rho\right)$$
$$-\exp\left(ikr+i\delta-\frac{m^{\frac{1}{2}}}{\sqrt{2}\hbar}\int_0^r\frac{\sigma(\rho)}{(E-V(\rho))^{\frac{1}{2}}}d\rho\right), \quad (63a)$$

where δ is the phase shift. Taking $\sigma=\sigma_0$ for $r\leqslant R$ and $\sigma=0$ for $r>R$, and taking for V the pure centrifugal potential $\hbar^2l(l+1)/2mr^2$, we find

$$\zeta=1-\exp\left(-\frac{4\sigma_0}{\hbar v}(R^2-l^2\lambda\!\!\!^{-2})^{\frac{1}{2}}\right). \quad (63b)$$

Summing over all l from 0 to $R/\lambda\!\!\!^{-}$, the total cross section is then

$$\sigma_t=\pi R^2\left\{1-2\frac{1-e^{-x}(1+x)}{x^2}\right\} \quad (64)$$

with

$$x=4\sigma_0R/\hbar v. \quad (64a)$$

§10. Influence of Nuclear Potential

Although we have given arguments for the assumption that the average nuclear potential V_n is small compared with the absorption potential σ (beginning of §4), we shall investigate the influence on our results of a nonvanishing V_n, attractive or repulsive. We shall assume that V_n is proportional to σ, *viz.*

$$V_n=\alpha\sigma. \quad (65)$$

Then the solution (16) of the Schrödinger equation is to be replaced by

$$u=cH_{2ikb}^{(1)}(2(i-\alpha)^{\frac{1}{2}}x). \quad (65a)$$

For small argument x, this becomes [cf. Eq. (16b)]

$$u=x^p-e^{\pi ip}(i-\alpha)^{-p}x^{-p}p!/(-p)!, \quad (65b)$$

so that (17) is replaced by

$$u=e^{-ik(r-R)}-e^{ik(r-R)}$$
$$\times e^{-(\pi+2\text{ arc tan }\alpha)kb+2i\eta-ikb\log(1+\alpha^2)} \quad (66)$$

and the formation probability (18) by

$$\zeta=1-e^{-2(\pi+2\text{ arc tan }\alpha)kb}. \quad (66a)$$

The main change is the appearance of $\pi+2$ arc tan α, instead of π, in the exponent. For an attractive potential, $\alpha<0$ and therefore the sticking probability is *decreased*. On the other hand, a repulsive nuclear potential will serve to increase ζ. The change of ζ will be only slight if α is small, i.e. if the nuclear potential is smaller than the absorption potential. Only for $|\alpha|\gg1$ and α negative, will the formation probability be very much less than for $\alpha=0$. Then we have practically only an attractive potential, and we consequently get more elastic scattering and less absorption. A large repulsive potential, on the other hand, increases the sticking probability for slow neutrons by a factor of two. Neither case is of practical importance.

The elastic scattering is modified by the term $-ikb\log(1+\alpha^2)$ in the exponential in (65) which corresponds to an increase of the radius of the effective hard sphere by $\frac{1}{2}b\log(1+\alpha^2)$, both for repulsive and attractive potential.

It is a pleasure to thank Dr. G. Placzek and Dr. V. F. Weisskopf for valuable discussions and suggestions.

Optical Activity, Circular Dichroism, and Absorption of Crystalline Nickel Sulphate

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(Received April 6, 1940)

Previous measurements of the optical rotatory power of crystalline $\alpha-NiSO_4 \cdot 6H_2O$ in the visible spectrum have now been extended to cover the wave-length range from 0.25μ to 2.2μ. Circular dichroism and absorption measurements have also been made over the infra-red range. Striking anomalies in the rotatory dispersion curve appear at 0.69μ and particularly at 1.16μ, corresponding to absorption and circular dichroism maxima at these wave-lengths. The theoretical relation between rotatory power and dichroism is verified quite exactly and in detail, particularly with regard to the 1.16μ band.

INTRODUCTION

THE optical activity of the crystalline alpha-hexahydrate of nickel sulphate has been measured for the visible[1] and near ultraviolet[2] spectral regions. The abnormal variation of the rotatory power with wave-length indicated that a major anomaly was to be expected in the infra-red.[3] This has been confirmed by the measurements here reported, which extend the rotatory power curve in both the infra-red[3] and ultraviolet to cover all the wave-length range from 0.25μ to 2.2μ. The vast majority of substances show a variation of rotatory power roughly as the inverse square of the wave-length. The relatively few materials which depart widely from this behavior, i.e., whose rotatory power is appreciably influenced by critical frequencies in the optical range, are of special interest. Nickel sulphate proves outstanding in this respect.

The property of circular dichroism, which is intimately associated with anomalous rotatory power, has also been measured for this material in the near infra-red.[4] This property consists of a difference in absorption for right-hand and left-hand circularly polarized radiation, and is most readily detected by the ellipticity which it produces in an incident plane polarized beam. Measurements of total absorption for the same infra-red region are also reported.

The infra-red measurements were made at the University of Wisconsin and those in the ultraviolet at Vanderbilt University. The two techniques are naturally different and will be described separately in the next two sections. In both cases the crystal specimens employed were laminae cleaved from Baker's or Merck's C.P. crystals, with thicknesses from 0.3 mm to 3.4 mm. These were examined by radiation transmitted normally and hence parallel to the optic axis, since the cleavage is along the 001 plane which is normal to the optic axis in the tetragonal crystal.[5]

METHOD AND APPARATUS—INFRA-RED

The apparatus for measuring optical rotation and the ellipticity of circular dichroism was a modification of that already described,[6-8] and need be discussed only briefly. Light from a special strip-filament tungsten lamp L (Fig. 1), after reflection from a concave mirror and passage through a Rochon double-image prism D_1 (only one beam used), was converged to a focus on the crystal C and then passed through a Wollaston double-image prism D_2. A concave mirror then formed two images with vertical and horizontal planes of polarization on the two slits (vertically in line) of a spectroradiometer having a differential vacuum bismuth-tellurium thermopile T with receiver strips 0.6×10 mm. The spectrometer had a 9-cm high 45° glass prism, which was transparent as far in the infra-red ($\lambda 2.4\mu$) as it has proved feasible to do

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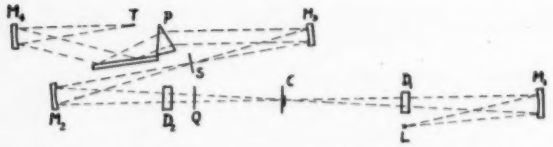

FIG. 1. Arrangement of apparatus for infra-red measurements.

polarization work of this sort. It was used in a Wadsworth mounting with mirrors of 40 cm focal length. For some of the work a higher dispersion was secured by replacing M_4 by a 60-cm mirror. All concave mirrors were aluminized. The spectrometer mirrors were of special Bausch and Lomb manufacture and their high quality was proved by Foucault knife-edge tests. The galvanometer was a Kipp and Sons type Zc.

The transmission of a crystal specimen could be measured at any wave-length by closing one slit completely with a shutter and observing galvanometer deflections with and without the crystal in the beam (the polarizing prisms having been removed), care being taken to keep the deflections within the limit of linearity, as determined by calibrated sector disks.

Figure 2 shows the method of measuring rotation. With the polarizer plane at azimuth 45°, as in (a), the two energy components falling on the differential thermopile are equal and the galvanometer shows no deflection. Insertion of the crystal will unbalance this condition, as in (b), because of rotation due to the crystal, but (c) turning the polarizer through an equal angle in the opposite sense will restore the balance. Actually it was found convenient to modify this ideal method to that illustrated in (d) and (e). When the polarizer was rotated 90°, x' changed to x'' (equal to y'), etc., with a corresponding change in the sign of the galvanometer deflection. By rotating the polarizer through the angle 2α, as in (e), the original galvanometer reading was restored.

The method of measuring ellipticities followed closely that already described.[8] Some 30 sheets of almost perfect optical mica, 5 cm square, were secured after much testing, which were quarter-wave plates for a series of wave-lengths from 0.6μ to 2.3μ. Since each plate could be used without appreciable error for a small spectral range on each side of the wave-length for which it was a true quarter-wave plate, it was possible with these plates to cover all wave-lengths of this region. The procedure was to mount a quarter-wave plate suitable for the desired wave-length at Q in a large conical bearing which permitted its rotation through exactly 90°. Such rotation turned the plane of the resultant plane polarized light through 2ϵ (ϵ, ellipticity), where $\tan \epsilon$ is the ratio of the axes of the ellipse.* This rotation was measured by comparing the resultant galvanometer deflection with that produced by a small (3°) known rotation of the polarizer D_1.

In measuring ellipticity due to circular dichroism it was not found possible to avoid additional ellipticity due to double refraction effects, such as would result from imperfect orientation of the crystal, or perhaps imperfections or strain in it. Ellipticity of such origin, however, must be reversed in sign if the polarizer is rotated through 90°, while ellipticity due to circular dichroism will remain unaltered. Measurements of ellipticity were therefore made for two orientations of the polarizer differing by 90° and their mean taken as the ellipticity due to dichroism alone. This procedure was tested by introducing deliberately a large double-refraction effect by rotating the crystal as much as 6° about a transverse axis. There was no change in the measured dichroism under these circumstances.

To secure sufficient energy for these spectroradiometric measurements it was necessary, as already mentioned, to use a conical beam, but to avoid birefringent effects it was necessary to keep the angle of the cone small, i.e., to deviate as little as possible from parallelism to the optic

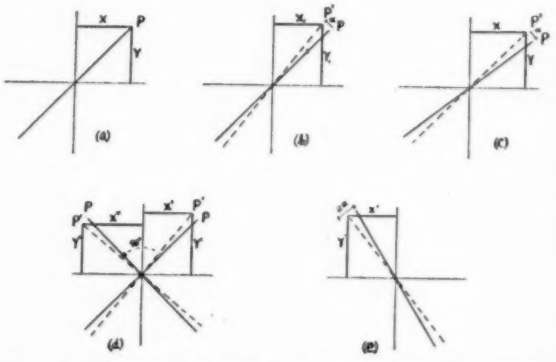

FIG. 2. Diagrams illustrating the method of measuring rotations. P is plane of polarization of the beam as it leaves the polarizer D_1; P' is plane after leaving crystal C.

* Reference 8, p. 412.

axis. The crystal lamina was mounted over a 5-mm aperture in a holder which allowed the crystal surface (001 plane) to be adjusted normal to the axis of the beam. A 15-mm or 25-mm diaphragm, located at Q, 50 cm from C (Fig. 1) limited the angle of the cone so that no part of the beam deviated more than 1.1° or 1.7°, respectively, from the optic axis. Measurements with the two diaphragms gave practically identical values for the cases of both rotation and ellipticity measurements, so it was concluded that no appreciable error resulted from lack of strict parallelism with the optic axis. In the case of the dichroism measurements, such errors would be eliminated anyway by the treatment of the data mentioned above.

Method and Apparatus—Ultraviolet

The arrangement of the apparatus used for measurement of rotations in the ultraviolet is shown schematically in Fig. 3. Radiation from the quartz mercury arc A passed through the Bausch and Lomb double prism monochromator B. The emergent monochromatic radiation then traversed successively the Glan-Thompson polarizer C, removable 45° glass plate D (used in orienting the crystal), the nickel sulphate crystal E, the Wollaston double-image prism F, and finally fell on the photographic film in the holder G, registering two narrow images of the monochromator exit slit. The dimensions of the apparatus were such that no ray deviated from the optical axis of the system by more than 1°.

It is obvious that the two photographic images produced by the Wollaston could be brought to equal density, by adjusting the plane of polarization of the incident radiation to an azimuth approximately 45° from the extinction points. This condition would be altered on insertion of the crystal, but the balance could be restored by rotating the polarizer to compensate for the rotation introduced by the specimen. This is illustrated in Fig. 2 (a), (b), (c). All photographs were measured with a microphotometer and the angles corresponding to equal densities of the two images were found by graphical interpolation. The amplifier used in the microphotometer employed the circuit described by Gabus and Pool[9] with a slight modification. A variable

[9] G. H. Gabus and M. L. Pool, Rev. Sci. Inst. **8**, 196 (1937).

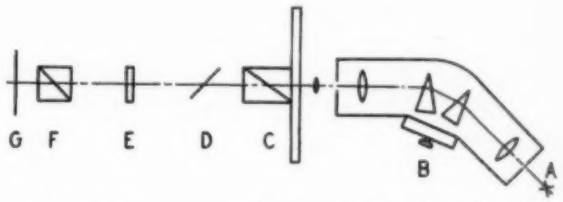

Fig. 3. Arrangement of apparatus for ultraviolet measurements.

bias was used on the control grid (actually the normal suppressor grid, as the grids are used in novel fashion), greatly facilitating the use of the amplifier and making it easier to adjust. The photo-cell (RCA type 922) was mounted over the eyepiece of the microscope, the usual illumination system being replaced by an assembly made for illuminating moving picture sound tracks.

To illustrate the method, the measurements obtained for the mercury line at 2804A are presented. Knowing approximately the zero points and rotations from previous trial photographs, four exposures, differing by −3°, −1°, +1°, and +3° from the expected correct position, were made both for the zero position and the rotated position in each quadrant—32 exposures in all for this wave-length. The relative density of the two images in each photograph was measured by placing the less dense image in the microscope field, and then adjusting the light until the galvanometer read 100. The more dense image was then immediately moved into the measuring position by means of the mechanical stage. Under these circumstances the galvanometer read directly the relative transmission of the denser image. Each set of the four relative transmission measurements is plotted graphically as shown in Fig. 4. Angular positions of the polarizer are abscissae and relative transmissions ordinates. The straight line through the four points intersects the line of equal transmissions, i.e., 100 percent, at the match point or angular position for equal density of the two images. Fig. 4 shows two such lines, one from data obtained with a 1.31-mm crystal in the optical path and the other with the crystal removed. The angular difference between the two match points is the rotation produced by the crystal. The value in this case is 39.8° −25° or 14.8°, which gives a rotatory power of

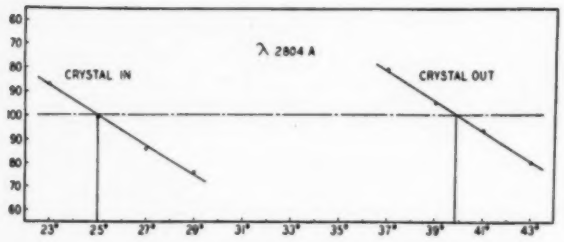

FIG. 4. Graph illustrating the method of determining rotations photographically. The abscissae are polarizer positions and the ordinates indicate relative transmissions in percent. Points are plotted above or below the mid-line depending on which of the two images is the denser.

11.3°/mm. This procedure was carried out for each of the wave-lengths used. The data shown in Fig. 4 are typical and indicate that the technique yields good results.

When the polarizer was set exactly 45° from the two planes of transmission of the analyzing Wollaston, the two images were not quite of equal density. This is at least in part caused by a difference in their size, and may also be a result of different absorption or reflection losses in the prism. Consequently, the match-points are not quite midway between the extinction points, and not quite 90° from one another. The effect is, however, common to the zero-point photographs and the rotation photographs, and causes no serious difficulty.

EXPERIMENTAL RESULTS

The measurements of rotation in the ultraviolet were made on two crystals, with respective thicknesses of 1.31 mm and 3.42 mm, the former giving right-handed rotations and the latter left-handed. The thicker specimen was used for most of the measurements except those near the absorption band around 3850A. Both crystals were used at 2804A, where the two values of the rotatory power agreed within 0.07°/mm. Measurements were made for a series of ultraviolet lines of the mercury arc extending from 2537A to 3650A. The results, expressed as rotatory power in degrees per millimeter, are given in the first section of Table I, and plotted as points in Fig. 5. A smooth curve has been drawn through the experimental points in this figure. Some of these data, and one additional measurement at 4047A, were also plotted in Fig. 6(a). No measurements between 3650A and 4047A have been possible because of an intense absorption band in this region. The break in the curve of Fig. 6(a) indicates the presence of this band and an associated rotational anomaly.

Four crystal specimens were used for the measurements in the infra-red, of thicknesses 0.33 mm, 0.64 mm, 0.83 mm, and 1.325 mm, obtained by micrometer caliper measurement. The 0.83 crystal gave positive rotations (clockwise to the observer) at the longest and shortest wave-lengths (i.e., where the rotation is "normal"

TABLE I. *Rotatory power, circular dichroism, and absorption coefficient of crystalline* $\alpha-NiSO_4\cdot 6H_2O$ *from 0.25μ to 2.2μ. The ultraviolet data are direct experimental values. The infra-red data are taken from the smooth curves in Fig. 6. The columns of differences were obtained by subtracting computed from experimental values.*

WAVE-LENGTH IN MICRONS	ROTATORY POWER IN DEGREES PER MM	DIFFERENCE EQ. (10)	CIRCULAR DICHROISM, DEGREES OF ELLIPTICITY PER MM	DIFFERENCE EQ. (9)	ABSORPTION COEFFICIENT PER MM
	Ultraviolet				
0.2537	18.4	−0.07			
0.2654	14.8	+0.03			
0.2804	11.4	−0.03			
0.2967	8.62	−0.2			
0.3130	6.92	−0.1			
0.3341	5.34	+0.1			
*0.3400	*4.9	+0.1			
*0.3500	*4.3	+0.2			
0.3650	3.11	−0.04			
0.4047	2.8	−0.05			
	Visible				
*0.43	*1.85	+0.1			*1.0
*0.45	*1.20	+0.04			*0.4
*0.50	*0.05	−0.07			*0.1
0.55	−0.9	−0.1			0.4
0.60	−1.85	−0.2	0.45	−0.02	1.8
	Infra-red				
0.63	−2.20 (max.)	−0.3	—	—	—
0.65	−2.15	−0.2	1.30	−0.04	3.4
0.675	—	—	—	—	3.5 (max.)
0.69	—	—	1.45 (max.)	+0.1	—
0.70	−1.75	−0.3	—	—	3.4
0.74	−1.65 (min.)	−0.3	—	—	—
0.75	—	—	1.05	−0.02	2.3
0.80	−2.25	−0.4	0.45	+0.07	1.1
0.85	−3.25	−0.5	0.35	−0.02	0.7
0.90	−4.60	−0.6	0.65	−0.1	0.9
0.95	−5.90	−0.4	1.65	−0.2	1.4
1.00	−6.95	−0.3	3.7	−0.02	2.1
1.02	−7.05 (max.)	−0.2	—	—	—
1.05	−6.70	−0.2	6.7	+0.2	2.7
1.10	−4.50	−0.2	9.7	+0.2	3.1
1.155	0.00	−0.1	11.5 (max.)	−0.2	3.3
1.175	—	—	—	—	3.4 (max.)
1.20	+3.50	+0.2	10.2	+0.2	3.3
1.25	5.55	−0.2	7.5	+0.4	3.0
1.30	—	—	4.9	+0.1	2.5
1.305	6.40 (max.)	−0.2	—	—	—
1.35	5.90	−0.2	2.9	−0.1	2.0
1.40	5.10	−0.04	1.70	−0.3	1.9
1.45	4.25	+0.04	1.00	−0.3	2.3
1.50	3.45	+0.02	0.55	−0.2	2.5 (max.)
1.55	—	—	0.25	−0.1	—
1.60	2.35	−0.1	0.10	−0.03	2.2
1.70	1.80	−0.1	—	—	2.0
1.80	1.40	−0.1	—	—	1.5
1.90	1.15	−0.1	—	—	3.2
1.98	—	—			8.8 (max.)
2.00	0.95	−0.1			—
2.10	0.80	−0.1			4.7
2.20	0.65	−0.2			2.9

* These data were taken from previous publications, by Slack and Rudnick, reference 2, or by Underwood, Slack and Nelson, reference 1.

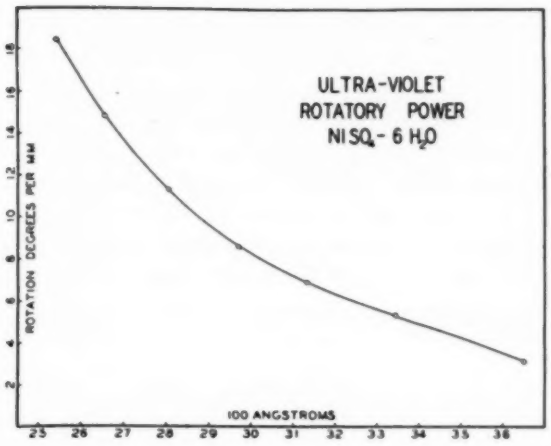

FIG. 5. Rotatory power of $\alpha-NiSO_4 \cdot 6H_2O$ in the ultraviolet.

in sign); the other three crystals were opposite in sign. Since no detectable difference in magnitude appeared in the rotatory powers derived from the two types of crystal, no attention was given to absolute sign in combining the data.

While temperature is a secondary factor in this work and was not under special control room temperature did not vary greatly from 24°C during these measurements and may be taken to have that value.

In selecting data for the final determination of rotatory power, no individual readings were rejected, but certain groups of measurements were omitted from the final plotting, largely as a matter of convenience. These were data taken with a filter or with the small diaphragm previously mentioned, which were not significantly different from the other results and at the same time were naturally a little less self-consistent because of the reduced intensity of radiation. The measurements which were retained were all obtained with the larger (25 mm) diaphragm. As mentioned before, a 40-cm focus mirror was employed at M_4 (Fig. 1) in the greater part of the work. This gave an effective slit width ranging from 0.02μ at wave-length 0.6μ to 0.09μ at 1.8μ. Some measurements with higher resolving power were obtained by use of the 60-cm mirror at M_4. The range of wave-lengths extended from 0.6μ to 2.2μ.

The observations (230 in number) remaining after the above-mentioned exclusion were expressed as rotatory power in degrees per millimeter and plotted; the smooth curve shown in Fig. 6(a) was then drawn through them. It was not feasible to show all of the experimental points or the full extent of their self-consistency

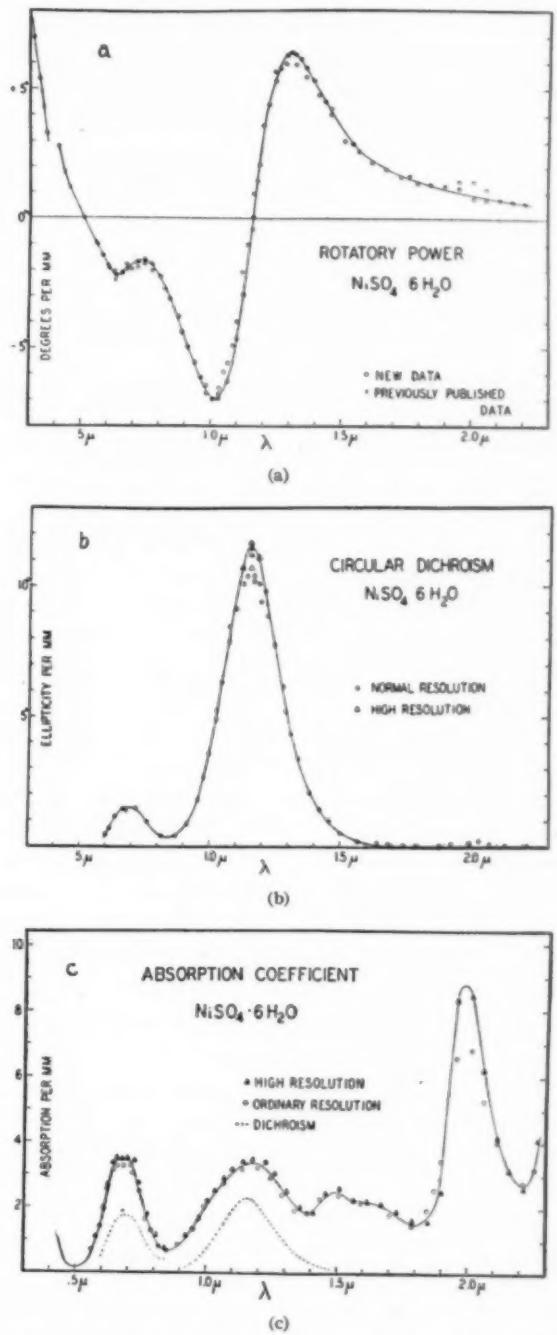

FIG. 6. Optical properties of $\alpha-NiSO_4 \cdot 6H_2O$ in the visible and infra-red. (a) Rotatory power. (b) Circular dichroism. (c) Absorption coefficient. Data shown in (a) and (c) for the visible region are taken from previous publications (references 1 and 2).

in this small figure. Hence only a small number of the experimental points are actually shown, but these include the points departing most from the smooth curve, thus indicating the extreme limits of variation among the measurements. The deviations from the curve near the peaks (approximate wave-lengths 0.7μ, 1.0μ, and 1.3μ) arise mainly from the differences in resolving power; higher resolving power gave slightly sharper peaks which were of course preferred in drawing the smooth curve. The scatter near 2.0μ reflects the difference between data from thick and thin crystals, the latter giving lower values. Values of the rotatory power read from the smooth curve appear in Table I. The wave-lengths of the peaks of the curve are indicated in the table as exactly as they could be determined.

Circular dichroism was measured over the same (infra-red) wave-length range in the two crystals of thickness 0.33 mm and 0.83 mm, which were of opposite sign. The larger diaphragm (25 mm) was used throughout; the lower resolving power was used except for a few measurements in the neighborhood of the principal maxima. The sign of the dichroism agreed, as expected, with that of the rotation at longer wave-lengths, and was ignored in combining the data. All measurements (101 in number), without any rejections, were reduced to ellipticity in degrees per millimeter of thickness of a very thin crystal,[10] then were plotted and through them was drawn the smooth curve of Fig. 6(b). Here again the small figure cannot show all the points or their full self-consistency, but the extreme divergences are shown. In particular, the scattered high dispersion measurements at the peak (1.155μ) were derived from a thick crystal which transmitted little radiation at that wave-length, while the peak of the smooth curve was fixed by three measurements on the thinner crystal which agreed with one another within 1 percent. The values of dichroism entered in Table I were read from the smooth curve.

The precision of the infra-red measurements appears to be limited by the finite resolving power used, and possibly by other sources of small systematic errors, since the small systematic differences for crystals of different thickness, or for different resolving powers, exceeded the random errors during any one run, except where the intensity of radiation was least. We believe the values in Table I for the infra-red region, both of rotatory power and ellipticity, have a probable error of 0.1°/mm or at most 0.2°/mm. The uncertainty is perhaps a little greater at the sharpest peaks of both curves, which may still be a little rounded by finite resolution, and also at the very steep places on both curves, where only a very slight revision of the assigned wave-lengths would be required to produce a change of 0.2°/mm in rotation or ellipticity at a given wave-length. The scale of infra-red wave-lengths was checked at several well-separated points and is believed dependable to about 0.01μ. The probable error of the ultraviolet measurements is considered also to be within 0.2°/mm.

Transmission was measured as a function of wave-length over the same infra-red range covered by the rotation and dichroism measurements, for three of the crystals (0.33 mm, 0.64 mm, 0.83 mm thickness). Absorption coefficients per millimeter were computed, assuming 4 percent reflection loss at each surface, and plotted in Fig. 6(c). The values from the crystals of different thickness agree well except in the very intense absorption band at 2.0μ, where a small amount of scattered light greatly lowers the apparent absorption of the thicker crystals and probably affects even the data from the thinnest crystal. Some systematic difference between the results with high and low resolving power appears in the wave-length region from 1.1μ to 1.8μ. A smooth curve has been drawn in Fig. 6(c) which compromises these differences and follows the points from the thinnest crystal near 2.0μ.

Discussion of Results

The carriers of all the absorption bands shown in Fig. 6(c) are known with at least reasonable

[10] In treating the ellipticity data two approximations were made for which corrections were later applied where necessary. The first was that galvanometer deflections were assumed proportional to angles of rotation equal to the ellipticity ϵ, and to $(\epsilon-3°)$, where the 3° is a rotation introduced for calibration purposes. More strictly, the deflections are proportional to $\sin 2\epsilon$ and $\sin 2(\epsilon-3°)$. The second approximation was that the ellipticity was proportional to the thickness of the specimen, whereas it actually approaches 45° as a limiting value for great thicknesses. First-order correction for these two approximations was made by multiplying by the respective factors $(1-4/3\epsilon^2)$ and $(1+\frac{2}{3}\epsilon^2)$, which combine conveniently to $(1-\frac{2}{3}\epsilon^2)$, where ϵ is the ellipticity actually measured, expressed in radians.

certainty. The bands with centers at 0.385μ and 0.69μ together produce the green color characteristic of divalent nickel salts in aqueous solution or in hydrated crystals, and are to be attributed to the hydrated nickel ion.[11] The band at 1.16μ is believed to belong to the same system, corresponding to the band at 1.2μ observed by Houstoun[11] for the aqueous solutions of six nickel salts. These three bands contribute to the rotatory power and are therefore presumably electronic in character.

The two bands which center near 1.5μ and 2.0μ have been identified as due to water of crystallization, by their close correspondence in position and absolute intensity with known bands of liquid water.[12] They are vibrational in character and, as expected, make no appreciable contribution to the rotatory power. The experimental data, both for dichroism and rotatory power, show a little suggestion of anomaly within the very intense band at 2.0μ, but we interpret this as an effect of scattered radiation, since it was least noticeable for the thinnest crystal, whose transmission was 10 percent at this wave-length.

Since electronic bands do not usually occur in the infra-red region, anomalous rotatory dispersion in the infra-red is correspondingly unusual. The anomaly at 1.16μ, is, furthermore, because of its favorable position, remarkably well isolated from the "normal" rotatory dispersion controlled by the far ultraviolet absorption. At the center of this band, the contributions to rotatory power by all other bands in the spectrum can hardly exceed 0.7°/mm, or 10 percent of the greatest rotations in this region. We know of few other instances where one anomaly is so completely isolated. This band therefore affords a highly favorable opportunity for quantitative study of the associated rotatory dispersion and dichroism.

Circular dichroism consists of a difference in absorption coefficients for the two senses of circularly polarized light, and rotatory power arises from a corresponding difference in refractive indices. It is to be expected that difference in absorption and difference in refraction will be related in much the same way as are total absorption and refraction, and this is qualitatively true. A narrow absorption line showing circular dichroism will make a contribution to the rotatory power at any wave-length not too remote from it. The variation of this contribution with wave-length resembles an anomalous dispersion curve, the contribution outside of the line itself being of the form $A/(\lambda^2-\lambda_0^2)$ or $B\nu^2/(\nu_0^2-\nu^2)$. The magnitude and sign of A and B are completely fixed by the dichroism of the line. If $d\alpha$ is the contribution to rotatory power at frequency ν made by an absorption line at frequency ν_1, ϵ the ellipticity produced by the dichroism per unit length for short paths, and $d\nu_1$ the effective width of the line (more exactly, $\epsilon d\nu_1$ is the area under the curve of ϵ against ν_1 within the line), then[13]

$$d\alpha=\frac{2}{\pi}\frac{\nu^2}{\nu_1^2-\nu^2}\frac{\epsilon d\nu_1}{\nu_1}. \tag{1}$$

Kuhn and Braun[14] have integrated this equation for the case of a broad absorption band within which the total absorption coefficient is a Gaussian probability function on a frequency scale. Such a broad band is regarded as a composite of many narrow lines, and the ratio of dichroism to total absorption for these lines is taken proportional to their frequency. The following relations then hold:

$$\tfrac{1}{2}(a_l+a_r)=a=a_0\exp\left[-\{(\nu_0-\nu)/\theta_\nu\}^2\right], \tag{2}$$

$$\tfrac{1}{4}(a_l-a_r)=\epsilon=\epsilon_0\frac{\nu}{\nu_0}\exp\left[-\left(\frac{\nu_0-\nu}{\theta_\nu}\right)^2\right], \tag{3}$$

$$\alpha=\frac{2}{\pi^{\frac{1}{2}}}\epsilon_0\frac{\nu}{\nu_0}\left[J\left(\frac{\nu_0-\nu}{\theta_\nu}\right)-\frac{\theta_\nu}{2(\nu_0+\nu)}\right]$$

$$\equiv\alpha_\nu(\epsilon_0,\ \nu_0,\ \theta_\nu), \tag{4}$$

$$\cong\frac{2}{\pi^{\frac{1}{2}}}\epsilon_0\theta_\nu\lambda_0^3\frac{1}{\lambda^2-\lambda_0^2}. \tag{4'}$$

Here a is the total absorption coefficient for unpolarized light, a_l and a_r the coefficients for the two senses of circularly polarized light, ϵ

[11] R. A. Houstoun, Proc. Roy. Soc. Edinburgh **31**, 538 (1911); also R. A. Houstoun and J. S. Anderson, *ibid.*, **31**, 547 (1911).

[12] Int. Crit. Tab. **5**, 269 (1929).

[13] See T. M. Lowry, *Optical Rotatory Power* (Longmans, Green), Chap. XXXIV.

[14] W. Kuhn and E. Braun, Zeits. f. physik. Chemie **B8**, 281 (1930).

ellipticity as above, and α the partial rotatory power contributed by the whole band. ν_0 is the frequency of the maximum of total absorption, and a_0 and ϵ_0 the respective values of a and ϵ at that frequency, while θ_ν is a constant determining the width of the absorption band. λ_0 is the wave-length corresponding to the frequency ν_0. $J(x)$ is a function given by

$$J(x)=e^{-x^2}\int_0^x e^{y^2}dy \tag{5}$$

$$\leqq x-(2/3)x^3+\cdots \quad \text{for } x \text{ small} \tag{5'}$$

$$\leqq 1/(2x)+1/(4x^2)+\cdots \quad \text{for } x \text{ large.} \tag{5''}$$

This function controls the rotatory power within the band, being zero at $x=0$, assuming its greatest positive and negative values when x is, respectively, about $+0.9$ and -0.9, and falling back to zero at $x=\pm\infty$. Kuhn[15] has published numerical values of J, and the approximate values given by (5′) and (5″) are also useful for $x<\frac{1}{2}$ or >3. The expression (4′) for rotatory power is valid at sufficient distances from the absorption band, and is derived from (4) by the use of the first term of the asymptotic expansion (5″). In (4′), θ_ν must be expressed as a wave number.

Lowry and Hudson[16] found that the corresponding relations for a band in which the total absorption curve was Gaussian on a wave-length scale involve the same function $J(x)$. In this case

$$a=a_0\exp\left[-\{(\lambda-\lambda_0)/\theta_\lambda\}^2\right], \tag{6}$$

$$\epsilon=\epsilon_0\frac{\lambda_0}{\lambda}\exp\left[-\left(\frac{\lambda-\lambda_0}{\theta_\lambda}\right)^2\right], \tag{7}$$

$$\alpha=\frac{2}{\pi^{\frac{1}{2}}}\epsilon_0\frac{\lambda_0}{\lambda}\left[J\left(\frac{\lambda-\lambda_0}{\theta_\lambda}\right)+\frac{\theta_\lambda}{2(\lambda+\lambda_0)}\right]$$

$$\equiv\alpha_\lambda(\epsilon_0,\lambda_0,\theta_\lambda) \tag{8}$$

$$\leqq\frac{2}{\pi^{\frac{1}{2}}}\epsilon_0\theta_\lambda\lambda_0\frac{1}{\lambda^2-\lambda_0^2}. \tag{8'}$$

When Eq. (4), (4′), (8), or (8′) is used over a sufficiently wide spectral range, the right member of the equation should contain an additional factor $(n^2+2)/(n_0^2+2)$, where n and n_0 are indices of refraction at respective frequencies ν and ν_0 (or wave-lengths λ and λ_0).

Circular dichroism should determine the associated partial rotatory power completely and uniquely through Eq. (1), which contains no adjustable constants. Our data have been used to test this relation in the following way: First expressions of the type of (3) and (7) were combined to give the best realizable numerical representation of the dichroism. The corresponding values of partial rotatory power were then computed from (4) and (8) and subtracted from the experimental values. The resulting differences, presumably the contribution of the far ultraviolet absorption, proved to be reasonably well, though not perfectly, representable by two terms of the Drude type (Eq. (4′) or (8′)). Eqs. (9) and (10) are the outcome of this procedure.

$$\epsilon=\frac{1.165}{\lambda}\left\{6.95\exp\left[-\left(\frac{\lambda-1.165}{0.151}\right)^2\right]+4.27\exp\left[-\left(\frac{0.858-\nu}{0.120}\right)^2\right]\right\}$$
$$+\frac{0.695}{\lambda}1.545\exp\left[-\left(\frac{1.44-\nu}{0.192}\right)^2\right], \tag{9}$$

$$\alpha=\frac{n^2+2}{n_0^2+2}\Big[\alpha_\lambda(6.95,1.165,0.151)+\alpha_\nu(4.27,0.858,0.120)+\alpha_\nu(1.545,1.44,0.192)$$
$$+\frac{0.009}{\lambda^2-0.150}+\frac{0.575}{\lambda^2-0.035}\Big]. \tag{10}$$

In these equations, ellipticities and rotations are expressed in degrees per millimeter, wave-lengths in microns, and frequencies are replaced by wave numbers in reciprocal microns. Numerical values of the fraction containing the indices of refraction in Eq. (10) were obtained from a previously published dispersion formula.[2] They

[15] W. Kuhn, Freudenberg's *Stereochemie*, p. 381. See also reference 13.

[16] T. M. Lowry and H. Hudson, Phil. Trans. Roy. Soc. **232**, 117 (1933).

ranged from 1.06 at 0.25μ to 1.01 at 0.5μ, the values being practically 1.00 for all longer wave-lengths. The differences between the values of rotatory power and dichroism given by Eqs. (10) and (9) and the experimental data appear in the third and fifth columns of Table I. It may be noted that the dichroism near 0.69μ is represented by Eq. (3), while that near 1.16μ is intermediate in character between (3) and (7) and is represented by a linear combination of bands of the two types with their centers coincident. Eq. (9) yields a peak at 0.69μ which is a little too sharp and one at 1.155μ which is not quite sharp enough, but on the whole it represents the observed dichroism without serious error. The corresponding values of rotatory power, from Eq. (10), reproduce the observations practically within the experimental uncertainties everywhere except in the spectral region from 0.6μ to 1.0μ. The discrepancy in this range, however, varies quite smoothly and gradually with wave-length. The maximum disagreement of 0.6°/mm at 0.9μ, and the irregularity at 1.2μ both fall at steep places on the curve, where they might be removed by comparatively small shifts along the wave-length axis. *No trace of the maxima and minima of the total rotation curve appears in the residual differences;* this result is regarded as a sensitive and successful test of the theoretical relation (1) between circular dichroism and partial rotatory power.

The ratio $(a_l-a_r)/a$ is called by Lowry[17] a dissymmetry factor. Its value at the center of the 1.16μ band is 0.24, which is unusually high; in the 0.69μ band the ratio is only 0.03. This factor is expected to be proportional to frequency within any one band; this implies that the maximum of dichroism will occur at a slightly higher frequency than the maximum of total absorption. This appears to be the case in the 1.16μ band, but both dichroism peaks are narrower than those of total absorption, and the proportionality with frequency is not quantitatively borne out.

In a review of the theory of optical activity Condon[18] has defined a quantity R, called rotational strength, which is a property of a single molecule associated with one transition in its absorption spectrum and measures the magnitude and sign of the dichroism and rotatory power contributed by that transition. A closely analogous quantity S, called the line strength, measures the contribution of the same transition to total absorption and refractivity. For the 1.16μ band, $R=1.7\times10^{-40}$, and $S=3.2\times10^{-39}$ if determined from the dichroism curve and the dissymmetry factor 0.24 (or 4.3×10^{-39} may be estimated from the total absorption). From quantum-mechanical theory one may take these two quantities to have respective orders of magnitude pm and p^2, where p and m are the magnitudes of the matrix elements associated with the transition in question in the respective matrices of electric and magnetic dipole moment, even though the explicit expressions given by Condon are for random orientation. It follows that in this case p is of the order of 0.02, in terms of the atomic unit equal to the dipole moment of the hydrogen atom in its normal state, and m is of the order of 0.3 Bohr magneton. This band therefore falls in the class mentioned by Condon for which the rotatory power exists by virtue of a strong magnetic moment and a relatively weak electric moment associated with the transition.

We wish to express our indebtedness to Francis J. Davis for his assistance in certain of the infra-red measurements, and to Clifford K. Beck and John J. O'Connor for making some of the ultraviolet measurements, also to Wilson G. Puryear for his participation in the latter work. We also wish to acknowledge our appreciation of assistance from the Wisconsin Alumni Research Foundation in connection with the infra-red work.

[17] T. M. Lowry, *Optical Rotatory Power*, p. 394.

[18] E. U. Condon, Rev. Mod. Phys. **9**, 432 (1937).

On a Proposed Thermoelectric Origin of the Earth's Magnetism

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(Received April 8, 1940)

It has been suggested that the earth's magnetism might arise from thermoelectric forces within the earth, the asymmetrical thermal distribution being due to material convection guided by Coriolis forces from the earth's rotation. The observed heat flow through the crust so limits the possible velocities and temperature differences that the Coriolis force seems to be inadequate by about a factor 10^6.

IN the face of the well-known difficulty of obtaining a theory to account for the order of magnitude of the earth's magnetic moment, 10^{26} dyne$^{\frac{1}{2}}$ cm^2, based on the known laws of physics, Elsasser has made an ingenious proposal.[1] He suggests that the convective currents within the earth, such as postulated by geophysicists in connection with continental drift and similar problems, are guided by Coriolis forces due to the earth's rotation. This would provide a thermal distribution sufficiently asymmetric to give rise to thermoelectric currents which would make a magnetic dipole moment (as well as moments of higher order). We propose to show, however, that the limitation upon the temperature differences and velocities, imposed by the known flow of heat through the crust, does not permit both the Coriolis forces and the temperature differences to be large enough for the purpose (unless matter within the earth behaves in a way radically different from that anticipated by extrapolating from laboratory pressures).

In the case most favorable to a large magnetic moment, the convection currents would have something like cylindrical form with their axes parallel to the axis of rotation. The length of the cylinders would be approximately R, the radius of the earth (or of the core), and we may consider that there are about six cylinders, each of diameter R. Let the temperature difference between the upward and downward part of such a current be ΔT. We may assume that the thermoelectric power, Q, of such a circuit has the order of magnitude known in ordinary laboratory thermocouples, say $Q=10^{-3}$ volt deg.$^{-1}$. It does not seem plausible to us that Q could be nearly that large as a consequence of inhomogeneous distribution of different materials within the turbulent matter, as visualized in the detailed part of Elsasser's suggestions. Such values might be approached, however, because of the existence of a pressure couple within the vortex.[2] The resultant magnetic moment of the vortices is then about $\sigma Q\Delta T R^3$, where σ is the specific conductivity, say about 10^4 dyne$^{\frac{1}{2}}$ volt^{-1} cm^{-1}, as for ordinary iron. Since this is the magnetic moment in the most favorable case, we have the inequality

$$\sigma Q\Delta T R^3 = 2\times 10^{25}\Delta T \text{ dyne}^{\frac{1}{2}}\text{ cm}^2\text{ deg.}^{-1} \geqslant 10^{26}\text{ dyne}^{\frac{1}{2}}\text{ cm}^2$$

or

$$\Delta T \geqslant 5°\text{C}. \qquad (1)$$

Such a thermal convection is motivated by a driving force per unit volume which we may write $D=\alpha\rho g\Delta T$, where α is the volume coefficient of expansion, ρ the density, and g the acceleration due to gravity, which might average about 600 dynes per gram within the earth. We may safely take α about 10^{-3} deg.$^{-1}$. Using (1), we have for the average driving force per unit volume

$$D \geqslant 0.03\rho \text{ dyne g}^{-1}. \qquad (2)$$

The Coriolis force per unit volume is $C\approx 2\rho\omega v$, where v is the prevailing velocity of the convection and ω is the angular speed of the earth, 10^{-4} sec.$^{-1}$. The possible value of v is limited by the known flow of heat up through the crust

[1] W. M. Elsasser, Phys. Rev. **55**, 489 (1939).

[2] D. R. Inglis and E. Teller, Phys. Rev. **57**, 568A (1940).

of the earth, 5×10^{12} calories per second, much of which is due to radioactivity in the crust. The upward convective transport of heat within the earth we may write $R^2v\rho s\Delta T$, where s is the specific heat. Taking the reasonable value $\rho s=1$ cal. deg.$^{-1}$ cm^{-3}, we have the inequality

$$3\times10^{17}v\Delta T<5\times10^{12}\text{ deg. cm sec.}^{-1} \quad (3)$$

or, together with (1)

$$v<3\times10^{-6}\text{ cm sec.}^{-1} \quad (4)$$

Using this value of v, we have for the Coriolis force per unit volume

$$C<6\times10^{-9}\text{ dyne g}^{-1}. \quad (5)$$

This is very much smaller than the driving force per unit volume (2), which must be opposed by a comparable frictional drag in a steady state. It does not seem plausible that this very weak Coriolis force should have an appreciable influence in determining the shape of the convection currents.

Our considerations do not exclude the possibility of material convection guided by Coriolis forces. But such convection can be assumed only if ΔT is much less than 5°, and this would be insufficient to account for the earth's magnetism by any mechanism yet proposed.

JUNE 15, 1940 PHYSICAL REVIEW VOLUME 57

Discussion on the Isotopic Weight of C^{12}

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(Received July 28, 1939)

The precision measurements of the masses of the three substandards, especially that of C^{12} by different observers show a slight discrepancy well outside the probable error. The masses of all isotopes determined by the doublet method depend more or less on the masses of the substandards. Therefore Dr. Aston has expressed anxiety that some agreement should be reached on the isotopic weight of C^{12} to enable a satisfactory table to be adopted. In the present paper some of Aston's objections and criticisms are discussed in some detail as well as the checks which each author has to offer for his measurement of the principal doublet in question. At the end of the paper are given the masses of the three substandards H^1, D^2, C^{12} as well as the mass of N^{14} which are thought to be the most reliable at present.

IN several letters to *Nature*,[1] lastly[2] as chairman of the International Committee on Atoms, Dr. Aston has devoted his attention to the discrepancy between his mass-spectrographic determination[3] of C^{12}, and that of Bainbridge and Jordan[4] and my own.[5] The mass of C^{12} as well as that of H^1 and of D^2 are so important because at present they are used in the determination by the doublet method of all other isotopic masses. The masses of these three substandards are computed from the mass differences of the three principal doublets which Aston[6] first succeeded in measuring. The mass differences are given in Table I in terms of 10^{-4} unit of isotopic weight. For the determination of any other isotopic mass for example N^{14} there is needed in addition to these at least another doublet such as $(CH_2^{12}-N^{14})$.

TABLE I. *Mass differences in terms of 10^{-4} mass unit.*

DOUBLET	$(H_2^1-D^2)$	$(D_2^2-C^{12++})$	$(C^{12}H_4^1-O^{16})$	$(C^{12}H_2^1-N^{14})$
Mass number	2	6	16	14
Aston	15.2 ±0.4	423.6 ±1.8	360.1 ±1.6	124.5 ±0.7
Bainbridge and Jordan	15.3 ±0.4	421.9 ±0.5	364.9 ±0.8	127.4 ±1.1
Mattauch and Bönisch	15.39±0.021	422.39±0.21	364.06±0.40	125.81±0.23
Asada and others*			364.2 ±0.9	125.7 ±0.6

[1] F. W. Aston, Nature **137**, 357 (1936); **139**, 922 (1937); **141**, 1096 (1938).
[2] F. W. Aston, Nature **143**, 797 (1939).
[3] F. W. Aston, Proc. Roy. Soc. **A163**, 391 (1937).
[4] K. T. Bainbridge and E. B. Jordan, Phys. Rev. **49**, 883 (1936) abs. 123, 124; **51**, 384 (1937) abs. 59, 60; cf. M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 369 (1937).
[5] J. Mattauch, Zeits. f. tech. Physik **19**, 578 (1938) and Physik. Zeits. **39**, 892 (1938).
[6] F. W. Aston, Nature **135**, 54 (1935).
* T. Asada, T. Okuda, K. Ogata and S. Yoshimoto, Nature **143**, 797 (1939).

While Dr. Aston's value of the third principal doublet deviates somewhat from those of the others, the measurements by the different authors of the first two doublets agree quite satisfactorily. The discrepancy in the isotopic weight of C^{12} is based on the disagreement in the measurement of the third doublet. We will therefore concentrate our attention on this doublet, and investigate the reasons in favor of one or the other value.

For a better understanding I shall briefly discuss Aston's criticism as well as his objections to the use of the ($C^{12}H^1_4-O^{16}$) doublet—though one of his principal doublets—to determine the isotopic weight of carbon. He states[7] as follows:

"(1) It (the 16-doublet) is so wide (more than 22 units of packing fraction) that the effect of uncertainty in the absolute value of the dispersion coefficient may be serious. (2) The value (of C^{12}) obtained is dependent to the extent of about one-third on the isotopic weight of H^1. (3) The fact that one line is atomic and the other molecular may possibly cause asymmetry."

It is seen at once that all three objections hold as well for the second principal doublet ($D^2_3-C^{12++}$), which, however, has been measured with good agreement in all three mass spectrographs. It is as asymmetrical in Aston's sense as is the 16-doublet, and its width in units of packing fractions is 70. The value of C^{12} obtained from it is dependent in its whole extent on the isotopic weight of D^2 which is as good a substandard as H^1. The fact is that each new value of any of the three principal doublets alters each value of the three substandards (and with them all other isotopic weights). It is therefore not quite correct to compute the mass of C^{12} from a redetermination of the 16-doublet by using Aston's value of H^1 as T. Asada and co-workers have done recently. Their value for this doublet, which confirms the value of Bainbridge and Jordan and my own, influences somewhat the mass of H^1 (and of D^2). Therefore the Japanese authors either should have used our value of H^1, which is in agreement with their measurement, or, still better, they should have combined their value of the 16-doublet with any set of the other two principal doublets which, fortunately, show no discrepancy. The same holds true for their evaluation of the mass of N^{14} from their measurement of the doublet ($C^{12}H^1_2-N^{14}$). The resulting small alterations, however, are nearly within the limits of error, and in fact Aston's objection (2) is of little significance.

In his last letter to *Nature* Aston confines himself mainly to his objection (3). He investigates critically the reasons in favor of his working hypothesis, that, by some systematic error, asymmetrical doublets either are measured too low in his single focusing mass spectrograph or too high in the double focusing instruments. Since Aston believes he has confirmed his value of the 16-doublet by various checks (see below) he thinks that in particular two old doublet measurements of mine point to the second possibility. The first is the measurement of the doublet ($O^{16}H^1_2-O^{18}$) which is hard to obtain on account of the rarity of O^{18}, and for which I first found[8] the mass difference 125.7 ± 1.8; later measurements of H. Lichtblau[9] on somewhat better doublets gave 120, while Aston[10] obtains 104.4 ± 1.8. Until recently there were no measurements of nuclear energies available which admitted of a proof of this mass difference. Now, however, Burcham and Smith[11] have measured the reaction energies released by the bombardment of O^{18} and N^{15} by protons. From these and Aston's or my values of the principal doublets one computes for the mass difference ($O^{16}H^1_2-O^{18}$): 114.8 ± 3.6 or 113.3 ± 2.3. I therefore hope that by a redetermination of this doublet we shall meet halfway. The second is my measurement[12] of the doublets ($Si^{29}F^{19}_3-Sr^{86}$) and ($Si^{30}F^{19}_3-Sr^{87}$). These I had on only one exposure and that was admittedly deficient. As it turned out later, my values for the packing fraction of these two Sr isotopes fell below Dempster's[13] new curve for the heavy elements. As Flügge and v. Droste[14] have pointed out, a

[7] F. W. Aston, Nature **139**, 922 (1937).

[8] J. Mattauch, Phys. Rev. **50**, 671 (1936).

[9] J. Mattauch, Zeits. f. tech. Physik **18**, 526 (1937); Physik. Zeits. **38**, 951 (1937).

[10] F. W. Aston, Proc. Roy. Soc. **A163**, 391 (1937).

[11] W. E. Burcham and C. L. Smith, Nature **143**, 795 (1939).

[12] J. Mattauch, Naturwiss. **25**, 156 (1937).

[13] A. J. Dempster, Phys. Rev. **53**, 64 and 869 (1938).

[14] S. Flügge and G. v. Droste, Zeits. f. physik. Chemie **B42**, 274 (1939).

low value is quite feasible in this case in consideration of Hahn and Strassmann's[15] new discovery of the fission of the uranium nucleus. The packing fraction curve is in detail certainly not quite a smooth one, and it is highly probable that, with the rearrangement of the nuclear particles after the fission, preferably those nuclei are built up whose packing fraction lies below those of the neighbor elements. It is of interest that in one fission process Sr has been found[16] as one primary product of the fission. After all I do not wish to press this old measurement of single doublets. It, as well as that of ($O^{16}H^1_2-O^{18}$), belongs to the first measurements that I took with the mass spectrograph set up in collaboration with Dr. Herzog. Since then (and especially after the provisional measurements with Dr. Herzog[17]) numerous changes, some of them most incisive, were carried out.[18] The different stages are marked by alterations in the width of the slit. This was 0.08 mm in the old measurements (0.02 mm in the provisional determinations in collaboration with Dr. Herzog) and less than 0.01 mm in the last precision measurements. I think in the discussion as to the best value of the 16-doublet we are as justified in neglecting these old measurements as is Dr. Aston in omitting his first measurements which gave the high value 374 for the mass difference of this doublet. Since that time Aston has replaced numerous parts of his instruments just as we have done. If we also neglect the first value of Bainbridge and Jordan of this doublet, i.e., 369, I think we put the following discussion of the checks which each author has for his measurements on a more reliable basis.

According to his working hypothesis Aston seems to look upon the measurement of the symmetrical doublet ($C^{12}_2H^1_4-C^{12}O^{16}$) as the best check of his 16-doublet. This heavier doublet gives the same main difference because each component is 12 units higher. A computation from his data given without error yields the value 359.4. It is, however, not easy to avoid entirely traces of nitrogen. To separate the line N^{14}_2 from that of $C^{12}O^{16}$ a resolving power of at least 1 : 2500 is necessary, whereas Aston states[19] that his "resolving power is only sufficient to separate clearly lines differing by 1 in 2000." A slight trace of N_2 would therefore shift the center of the line towards $C^{12}_2H^1_4$, tending to lower the value of the doublet. Aston obtains another check by the determination of the doublets ($C^{12}_3H^1_4-A^{40}$), ($N^{20}-A^{40++}$), and ($O^{16}D^2_2-Ne^{20}$). These together with his value for the first two doublets give 361.2 ± 6.0 for the mass difference of the 16-doublet. The large error, however, does not allow a decision between the two values in question. The best check in fact is furnished by a combination of the doublet ($C^{12}_2-Ti^{48++}$) measured by Aston[20] and of the value for ($O^{16}-Ti^{48+++}$) obtained by Dempster.[21] From these with the value of Aston's first two principal doublets one obtains 360.5 ± 3.2 for the third. Though the large error does not exclude my value, Aston has given here a valuable hint for further measurements. The precision measurement of the two doublets containing Ti would reduce the number of principal doublets as well as the number of substandards to two, i.e., C^{12} and Ti^{48}. Most direct indeed would be the determination of the doublet ($C^{12+++}-O^{16++++}$), the triple value of which would give at once the deviation of the isotopic weight of C^{12} from the mass number. Thus we should have only one principal doublet and one substandard. Oxygen ions of such high charge are at present, however, provided only by Dempster's[22] elegant method of producing ions. Furthermore a resolving power well above 1 : 3000 would be necessary for the measurement of this doublet.

Bainbridge and Jordan have as a check of the 16-doublet the sum of the separately measured doublets ($C^{12}H^1_2-N^{14}$) and ($N^{14}H^1_2-O^{16}$) which gives 364.3 ± 1.9. In addition, they are able to separate N^{14}_2 clearly from $C^{12}O^{16}$ and they have measured the mass difference. When combined with ($C^{12}H_2-N^{14}$) this gives the value 366.5 ± 3.0 for the 16-doublet. Both checks agree with their

[15] O. Hahn and F. Strassmann, Naturwiss. **27**, 11 and 89 (1939).
[16] O. Hahn and F. Strassmann, Naturwiss. **27**, 163 (1939).
[17] J. Mattauch and R. Herzog, Naturwiss. **25**, 747 (1937).
[18] A paper describing the constructional details of the instrument and its performance is in preparation.
[19] F. W. Aston, Proc. Roy. Soc. **A163**, 396 (1937).
[20] F. W. Aston, Nature **141**, 1096 (1938).
[21] A. J. Dempster, Phys. Rev. **53**, 68 (1938).
[22] A. J. Dempster, Rev. Sci. Inst. **7**, 46 (1935).

direct determination 364.9±0.8 within the error allowed by them. Then their measurements of the doublets ($B^{10}H^1_2-C^{12}$), ($B^{10}-Ne^{20++}$), and ($O^{16}D^2_2-Ne^{20}$), the mass differences of which they are able to check up by further independent doublets, furnish a new evaluation of the isotopic weight of C^{12} for which, however, they use the 16-doublet in question. By computing this mass difference from the doublets above and the values of Bainbridge and Jordan for the first two principal doublets, we obtain 358.5±5.4 (or 359.8 with weighted means of the three doublets above, and their respective checks). This value agrees much better with Aston's though on account of the large error it does not contradict their own value.

In my work[23] done in collaboration with A. Bönisch, we first have checked up our determination of the doublet ($C^{13}H^1_2-N^{14}$), which is asymmetrical in the sense of Aston, by the measurement of the two symmetrical doublets ($C^{13}H^1_3-N^{14}H^1$) and ($C^{13}H^1_4-N^{14}H^1_2$). These repeat the same mass difference, respectively, one and two mass units higher. We obtained for the latter the values 125.63±0.27 and 126.26±0.51 which even within the small errors given by us agree with the direct measurement 125.81±0.23. This by no means supports Aston's working hypothesis. We have checked our value of the 16-doublet as Bainbridge and Jordan have done by combination of ($C^{13}H^1_2-N^{14}$) with ($N^{14}H^1_2-O^{16}$) as well as with ($N^{14}_2-C^{12}O^{16}$) which gives 363.61±0.40 and 363.84±0.61. These values are to be compared with3 64.06±0.40, the result of the direct determination. Again the checks fit in even within our small errors.[24]

We are justified in measuring the narrow doublet ($N^{14}_2-C^{12}O^{16}$) because our separation of the far narrower doublets ($C^{12}O^{16}H^1-C^{13}O^{16}$) and ($C^{12}_2H^1_5-C^{12}C^{13}H^1_4$) both at mass number 29 proves experimentally that our resolving power is better than 1 : 6500.

Aston, of course, is perfectly right in warning against systematic errors. The search for them is always a tedious task but indispensable in precision measurements. We think, however, that our instrument makes the detection relatively easy and convincing[25] compared to the instruments of Aston, and of Bainbridge and Jordan. For we can easily compute the dispersion coefficient and its course—it is simply proportional to the distance of the line from a fixed point. It is easy to prove this relation experimentally. In order to convert the measured differences of length to mass differences we therefore do not need the knowledge of any particular mass or mass difference as is the case with other mass spectrographs. It is therefore easier for us to find systematic errors than it is in mass spectrographs of the type of Aston or of Bainbridge and Jordan, where the dispersion coefficient and its change along the plate must be found by calibration. Actually we find a very slight systematic change of the doublet width with the strength of the magnetic field. In other instruments it would not be as easy to find such a slight systematic change. It is probably caused by the stray field which cannot be shielded completely. The percentage error of our 16-doublet is therefore larger than that of the much wider 6-doublet which can be obtained by much smaller magnetic field strengths. This systematic error, by the way, tends to lower and not to raise the results of our measurements. It is, however, negligibly small.[26]

Another criticism of Aston's concerns the accuracy claimed. He states that my figure corresponds to an uncertainty of position of a line on the photographic plate of 0.0003 mm "which seems very small for photometric measurements." For the middle of my plate this figure corresponds to 0.4×10^{-4} unit of mass in the neighborhood of mass number 16. This is half the width of the probable error which I

[23] J. Mattauch, Zeits. f. tech. Physik **19**, 578 (1938) and Physik. Zeits. **39**, 892 (1938).

[24] Aston remarks: "The three doublets $CO-N^{14}_2$, $C-BH^{10}_3$ and $Ne^{20}-CD_4$ quoted by Mattauch to check his high value are useless in their evidence for it so happens that if my values for all the atoms concerned are put in the agreements are equally good." This, however, must be due to a misunderstanding. I have never measured the doublet ($B^{10}H^1_2-C^{12}$). I have not used the doublet ($C^{12}D^2_4-Ne^{20}$) to check up my value of the 16-doublet or of C^{12} nor could I have done so since I have not measured another doublet containing Ne^{20} which would permit its elimination. Aston's remark is correct for the doublet ($N^{14}_2-C^{12}O^{16}$) insofar as our discrepancies just cancel if one computes its mass difference from those of the 16- and 14-doublet. The doublet ($N_2{}^{14}-C^{12}O^{16}$), however, was never measured by Aston nor is he, as we have seen, able to separate it. Our measurement provides therefore an independent check of the 16-doublet.

[25] J. Mattauch, Naturwiss. **25**, 157 (1937); R. Herzog and V. Hauk, Zeits. f. Physik **108**, 609 (1938).

[26] A full account of our precision measurements and their results will be published elsewhere.

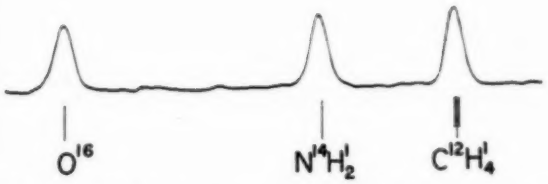

FIG. 1. Triplet at mass number 16. The hatched strip at $C^{12}H^1_4$ indicates the size of the probable error of a single measurement of doublet width. This in turn was the mean of the independent readings of three observers.

gave for the *mean* of all the 21 measurements of the 16-doublet. To make Aston's criticism applicable one has to use the probable error of a *single* measurement which is $21^{\frac{1}{2}}$ times larger. The whole width of it corresponds to an uncertainty of a line on the photographic plate of nearly 0.003 mm; this is after all one-fifth of the half-width of a line, a measurement which is easily possible. The photometric record of a typical 16-doublet together with the uncertainty corresponding to the width of the probable error of a single measurement is reproduced in Fig. 1. Again each single measurement was the mean of the independent readings of three observers. The distances of the lines were measured as usual with a comparator which permitted the direct reading of 0.001 mm, and the estimation of one-tenth of that amount. The photometric records were used only to establish the equality of the intensity of the lines.

It is usual with precision measurements to give the computed probable error which "defines the region for which the odds are even that the true value lies within it."[27] If systematic errors are found, they are to be included in such a way that the condition of even bet is preserved. I have always given the probable error and tried to avoid systematic errors as far as possible. I do not think it quite correct to enlarge this region of error needlessly as apparently has been done by Bainbridge and Jordan,[28] and which was formerly the habit of Aston. If this is done it renders the judgment of the checks difficult as we have seen, and it offers the author a rather unjustified chance that later more precise measurements will fall within his "limit of error." As it happens Aston's criticism is preceded by a "preliminary report on the masses of C^{12} and N^{14} by T. Asada and others. The authors report their measurements taken with a newly constructed mass spectrograph of the Bainbridge and Jordan type." These happen to be the measurements of the two doublets in question ($C^{12}H_4{}^1-O^{16}$), and ($C^{12}H^1_2-N^{14}$). (See Table I.) I appreciate very much the fact that their values fall within my small intervals of error as obviously they were obtained in perfect ignorance of my paper. This is made clear in that the Japanese authors, as mentioned above, used for the calculation of the isotopic masses of C^{12} and N^{14} Aston's value of H^1 which is not quite compatible with their own measurements.

I fully agree with the suggestion with which Aston concludes his criticism, that is to measure in the double focusing mass spectrograph other doublets suitable to check the value of C^{12}. Measurements of this kind are planned as soon as the mass spectrograph is in working order after its removal to Berlin. Until then, I think the values computed from our measurements of last year to be the most reliable. Meanwhile A. Bönisch has completed an evaluation by means of least squares which also takes care of the doublets used for checking, and the relations between them and the directly measured values. From this evaluation which gives a smaller error are obtained the following isotopic masses of the three substandards and of N^{14} which agree within the intervals of error with the values already published:

$$H^1 = 1.0081\ 30 \pm 0.0000\ 033$$
$$D^2 = 2.0147\ 22 \pm 0.0000\ 064$$
$$C^{12} = 12.0038\ 61 \pm 0.0000\ 24$$
$$N^{14} = 14.0075\ 3 \pm 0.0000\ 32.$$

[27] Cf. R. T. Birge, Phys. Rev. **52**, 241 (1937).

[28] Cf. p. 369 of the report of Livingston and Bethe (reference 4), where the latest results of Bainbridge and Jordan are published: "The errors are those given by Bainbridge and Jordan in cases (a) and (b) [i.e., the first and second principal doublet] while we have given a smaller error for CH_4-O. This doublet is based on 24 independent measurements, from 4 different plates, none of the measurements deviating more than 0.3 unit, so that 0.08 is more than twice the mean square error." [Here the values are given in terms of 10^{-3} mass unit, whereas I have used throughout this paper 10^{-4} unit of isotopic weight.] Since the probable error is equal to 0.6795 times the mean square error the error reduced by Livingston and Bethe is about 3 times the probable error corresponding to one chance in 22 that the deviation of the true value is as big as that. Where Livingston and Bethe have not reduced the error it seems to be still larger. Otherwise it is not to be understood why for example, for the first principal doublet Bainbridge and Jordan give an error nearly twenty times as big as the one we find, though the resolution of their mass spectrograph is at least as good as that of our instrument.

On the Establishment of Grand Canonical Distributions

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(Received December 22, 1939)

It is shown, in accordance with the accepted principles of statistical mechanics, that the establishment of a grand canonical distribution is to be expected in an ensemble which represents the attainment of equilibrium in an "open" system, connected with appropriate reservoirs for supplying the various kinds of molecules involved, and in thermal contact with a large heat bath at a given temperature. This finding is of assistance, in placing on a sound and understandable basis the use which has been made of grand canonical ensembles, for the representation of thermodynamic equilibrium in "open" systems.

§1. INTRODUCTION

IT is the task of the science of statistical mechanics to give satisfactory treatment to the properties and behavior of mechanical systems in situations where the condition of the system of interest is not sufficiently well determined to provide a complete specification of its precise mechanical state.[1] This task is performed by correlating the properties and behavior, to be expected for the system of interest itself, with the average properties and behavior for the members of a representative ensemble of systems of similar structure, which are appropriately distributed over different possible precise states. To obtain such an appropriate distribution, the members of the representative ensemble, in accordance with the fundamental postulate of statistical mechanics as to *a priori* probabilities and phases, are to be distributed with equal probabilities and random phases among the different possible precise states which agree equally well with the actual condition of the system of interest, insofar as that is determined.

In the case of systems, which from a microscopic point of view are composed of many separate molecules in molecular states that may be rapidly changing, but which from a macroscopic point of view are in a steady condition of equilibrium, three different kinds of representative ensemble have proved of special importance. These three kinds of ensemble are given the same names—microcanonical, canonical, and grand canonical—in a modern *quantum-mechanical* treatment, as were originally given to the analogous ensembles in the *classical* development of statistical mechanics by Gibbs.[2] Any ensemble of these three kinds can itself be shown to be in a condition of statistical equilibrium, and hence is thus in any case made suitable for representing a system of interest which for its part is in a condition of macroscopic equilibrium. The three kinds of ensemble differ from each other, however, with regard to the energies and compositions assigned to their component members, and are thus made suitable for representing systems of interest which have achieved equilibrium under different conditions of contact or connection with their surroundings.

In a *microcanonical ensemble*, each member of the ensemble is taken as having the same values for its external coordinates such as total volume, and for the numbers of constituent molecules of the various independent kinds present, as prevail in the system of interest itself; and the members are distributed over their different possible energy eigenstates, with random phases and with probabilities satisfying

$$\begin{aligned} P_i &= \text{const.} \quad && (E_i \text{ in range } E \text{ to } E+\delta E), \\ P_i &= 0 && (E_i \text{ not in range } E \text{ to } E+\delta E), \end{aligned} \tag{1.1}$$

[1] This holds both for the classical statistical mechanics, where the precise state of a system of f degrees of freedom would be specified by the exact values of $2f$ dynamical variables, and for the quantum-statistical mechanics, where the precise state of a system of f degrees of freedom would be specified by a probability amplitude for f dynamical variables. The treatment of statistical mechanics in this paper will be quantum-mechanical, since we can regard classical mechanics as a special limiting case of quantum mechanics.

[2] For the original classical description of these ensembles see Gibbs, *Elementary Principles of Statistical Mechanics* (New Haven, 1903). For a quantum-mechanical description, see for example, Tolman, *The Principles of Statistical Mechanics* (Oxford, 1938).

where P_i is the probability for finding a member of the ensemble in an eigenstate i corresponding to the energy eigenvalue E_i, and E to $E+\delta E$ denotes a small energy range which has been selected as of interest.

In agreement with the above-described character, a microcanonical ensemble may be regarded as representing a system which has been set up with precisely specified external coordinates such as total volume, with a precisely specified molecular constitution, and with its energy specified in a narrow range, and which has then been allowed to come to equilibrium under conditions of *perfect isolation* from the surroundings such as to preserve its total energy within the narrow range selected.[3] It will be appreciated that these requirements as to energy and isolation are not such as to make a microcanonical ensemble truly appropriate for the representation of thermodynamic equilibrium, since it would then be essential to regard the system as having a specified temperature determined for example by interaction with a heat bath, rather than to regard it as having a specified energy determined by the maintenance of perfect isolation.

In a *canonical ensemble*, each member of the ensemble is again taken as having the same values for its external coordinates and for its molecular constitution as prevail in the system of interest itself; but the members are now taken as distributed over their different possible energy eigenstates, with random phases and with probabilities satisfying

$$P_i = e^{(\psi - E_i)/\theta}, \tag{1.2}$$

where P_i is the probability for finding a member of the ensemble in an eigenstate i corresponding to the energy eigenvalue E_i, and ψ and θ are adjustable parameters having the dimensions of energy.

In agreement with the above-described character, a canonical ensemble may be regarded as representing a system which has been set up with precisely specified external coordinates and molecular constitution, and which has then been allowed to come to equilibrium under conditions of *interaction* with its surroundings such as to justify the probabilities for different energies described by (1.2). As first shown for the classical mechanics by Gibbs,[4] this interaction could be obtained by allowing the system to achieve *thermal equalization* with a large heat bath at temperature $T=\theta/k$, where interchange of energy between the system and bath would provide the fluctuations necessary to correspond to the above probability distribution in energy. Or, as shown more recently by the writer,[5] this interaction could also be obtained by leaving the system in *essential isolation*, where interchange of energy between the system and its containing walls, or other immediate surroundings, would provide the fluctuations necessary to correspond to the above probability distribution in energy, at least over a range in the neighborhood of the mean energy of the members of the ensemble, and where the resulting temperature $T=\theta/k$ would be determined by the mean energy, which in cases of essential isolation is taken as unchanged by the interaction permitted. It will be appreciated, as was first shown for the classical mechanics by Gibbs,[6] that the energy distribution in a canonical ensemble is such as to make it strictly appropriate for representing equilibrium in a "closed" thermodynamic system, where it would be essential to regard the system as having a precisely specified temperature rather than a precisely specified energy.

Finally in a *grand canonical ensemble*, each member of the ensemble is again taken as having the same values for its external coordinates as prevail in the system of interest itself; but the members are now taken as distributed both over different possible molecular constitutions, and over their different possible energy eigenstates, with random phases and with probabilities satisfying

$$P_{n_1, n_2, \cdots n_k, i} = e^{(\Omega + \mu_1 n_1 + \mu_2 n_2 + \cdots + \mu_k n_k - E_i)/\theta}, \tag{1.3}$$

[3] For a classical discussion of the process by which the representative ensemble for an isolated system, having a closely specified energy, proceeds in the direction of microcanonical distribution, see Lorentz, *Abhandlungen über theoretische Physik* (Teubner, 1907), §79, p. 289. For a quantum-mechanical discussion, see Tolman, reference 2, §§109 and 110.

[4] Gibbs, reference 2, p. 161. For the quantum-mechanical treatment of the same phenomenon, see Tolman, reference 2, §§111, 112 and 128.

[5] See in particular Tolman, reference 2, §111*d*.

[6] Gibbs, reference 2, Chap. 14. For the corresponding quantum-mechanical discussion, see Tolman, reference 2, Chap. 13.

where $P_{n_1, n_2 \cdots n_h, i}$ is the probability for finding a member of the ensemble with n_1, $n_2 \cdots n_h$ molecules of the h independent kinds present, and in an eigenstate i corresponding to the energy eigenvalue E_i, and where Ω, θ, and $\mu_1 \cdots \mu_h$ are adjustable parameters, having the dimensions of energy or of energy per molecule.

In agreement with the above-described character, a grand canonical ensemble may be regarded as representing a system which has been set up with precisely specified external coordinates such as total volume, and which has then been allowed to come to equilibrium under conditions of *interconnection and interaction* with its surroundings such as to justify the probabilities for different molecular constitutions and for different energies described by (1.3). The nature of the interconnection and interaction which would lead to this result has not hitherto been made the subject of direct investigation.

It is now the purpose of the present article to show that the conditions leading to the establishment of a grand canonical distribution are of such a character that the ensemble can be taken as representing a system of interest which has been allowed to come to equilibrium with its molecular constitution determined by interconnection with suitable reservoirs for providing the different kinds of molecules which it contains, and with its temperature determined by interaction with a suitable heat bath. This result will then be of assistance in placing on a sound and understandable basis the use which has been made of grand canonical ensembles, by Gibbs and others,[7] for the representation of equilibrium in so-called "open" thermodynamic systems.

§2. Distribution in Terms of Sum-Over-States for Reservoirs

In order to carry out the proposed demonstration, we shall wish to consider the equilibrium of a combined system composed of connected parts S, R_1, R_2, $\cdots R_h$. The part S, which may be called the system proper, will itself turn out to be the "open" system of primary interest for which we wish to derive the grand canonical distribution as representing equilibrium. It contains a mixture of all the different independent kinds of substance[8] 1, 2, $\cdots h$ that concern us. The parts R_1, R_2, $\cdots R_h$, which may be called reservoirs, are connected with S through semi-permeable membranes each of which allows the passage of a single kind of substance 1, 2, $\cdots h$, respectively. Each reservoir contains a supply of the substance for which its membrane is permeable. The volumes of the system proper and of the reservoirs are to be taken as having constant values, and for the purposes of the proposed treatment we may assume that external coordinates other than volume are not involved.

Let us now consider the equilibrium which would be reached if the above combination of system proper and reservoirs were allowed to stand in thermal contact with a large heat bath at temperature T. Since the combination as a whole is itself a "closed" thermodynamic system, its condition at equilibrium could be represented by the canonical distribution

$$P_i = e^{(\psi - E_i)/\theta}, \tag{2.1}$$

where P_i is the probability for finding a member of the ensemble in a state i corresponding to an eigenvalue of energy E_i for the combined system, where the parameter ψ has the necessary value so that the total probability for different states is normalized to unity, and where the parameter θ has the value

$$\theta = kT, \tag{2.2}$$

with k as Boltzmann's constant and T the temperature of the bath.

We may assume that the separation between the different parts of the combined system is sufficiently distinct, so that we can distinguish with accuracy between the molecules present in the system proper and those remaining in the reservoirs, and may assume that the interaction between the parts is sufficiently small so that we can regard any energy eigenstate of the whole system as a combination of energy eigenstates for its parts and regard the eigenenergy of the whole as the sum of the eigenenergies of its parts. This will then permit us to rewrite the description

[7] For the original classical application of grand canonical distributions to this purpose, see Gibbs, reference 2, Chap. 15. For the corresponding quantum-mechanical applications, see Tolman, reference 2, §140.

[8] The different "component substances," in the sense used by Gibbs in his treatise "On the equilibrium of homogeneous substances," Trans. Conn. Acad. III (1875–78).

of the distribution given by (2.1) in the more explicit form

$$P_{n_1, n_2, \cdots n_h, s, r_1, r_2, \cdots r_h} = \exp\left[(\psi - E_s - E_{r_1} - E_{r_2} - \cdots - E_{r_h})/\theta\right], \quad (2.3)$$

where we have replaced P_i by the more explicit symbol $P_{n_1, n_2, \cdots n_h, s, r_1, r_2, \cdots r_h}$, to denote the probability for a state of the combined system in which the system proper S contains n_1, n_2, $\cdots n_h$ molecules of the h kinds involved and is an eigenstate s, and in which the reservoirs R_1, R_2, $\cdots R_h$ containing the remaining molecules are in eigenstates $r_1, r_2, \cdots r_h$, and where we have replaced E_i by the sum of the eigenenergies E_s, E_{r_1}, E_{r_2}, $\cdots E_{r_h}$ for the parts of the whole system.

By summing the above expression for the probability of a state of the combined system over all states r_1, r_2, $\cdots r_h$ for the reservoirs, which are compatible with their molecular contents, we then obtain

$$P_{n_1, n_2, \cdots n_h, s} = e^{(\psi - E_s)/\theta} \sum_{r_1, r_2, \cdots r_h} \exp\left[-(E_{r_1} + E_{r_2} + \cdots + E_{r_h})/\theta\right]$$
$$= e^{(\psi - E_s)/\theta} \left(\sum_{r_1} \exp\left[-E_{r_1}/\theta\right]\right)\left(\sum_{r_2} \exp\left[-E_{r_2}/\theta\right) \cdots \left(\sum_{r_h} \exp\left[-E_{r_h}/\theta\right]\right) \quad (2.4)$$

as an expression for the probability of a state of the system proper, which is specified by its molecular constitution n_1, n_2, $\cdots n_h$ and by an eigenstate s possible for that constitution. It will be seen that this expression depends on the so-called sum-over-states for the system of reservoirs, and hence on the product of those quantities for the separate reservoirs, where it will be appreciated that the sum-over-states for each reservoir will itself depend, not only on volume and temperature which may be regarded as fixed, but also on the number of molecules in the reservoir, which will vary with the composition of the system proper.

To obtain a notation which indicates that the value for the sum-over-states of each reservoir is dependent on its molecular content, it will be convenient to retain the symbol n_i for the number of molecules of the ith kind, $i = 1, 2, \cdots h$, in the system proper S, and to introduce the new symbols n_i' for the number of molecules of the ith kind in the corresponding reservoir R_i, and n_i^0 for the constant total number in the combined system as a whole. This will give us

$$n_i' = n_i^0 - n_i \quad (2.5)$$

as the value for the number of molecules in the ith reservoir R_i. Ror the sum-over-states of the ith reservoir, we may then introduce the symbol

$$Z_{n_i'} = \sum_{r_i} \exp\left[-E_{r_i}/\theta\right], \quad (2.6)$$

where the subscript to Z indicates that the summation over possible states r_i is to be evaluated when the reservoir contains n_i' molecules.

Substituting symbols of the above form into (2.4), we can now write

$$P_{n_1, n_2, \cdots n_h, s} = e^{(\psi - E_s)/\theta} Z_{n_1'} Z_{n_2'} \cdots Z_{n_h'} \quad (2.7)$$

as our expression for the probability of a state of the system proper of the indicated composition and energy. By studying the dependence of the sums $Z_{n_1'}$, $Z_{n_2'}$, $\cdots Z_{n_h'}$ for the reservoirs on the numbers of molecules n_1', n_2', $\cdots n_h'$ which they contain, and hence in accordance with (2.5) on the numbers of molecules n_1, n_2, $\cdots n_h$ in the system proper, it will then prove possible to obtain the desired information as to the manner in which the probability for a state of an "open" thermodynamic system depends at equilibrium on its composition as well as on its energy.

§3. Lemma on the Dependence of Sum-over-States on Number of Molecules

The precise evaluation of the sum-over-states for a system of interacting molecules is in general a complicated and difficult task which often has to be solved by some method of approximation. It will, however, be possible to obtain the needed information as to the dependence of the sum-over-states of a system on the number of molecules which it contains by making use of the two following known relations. In the first place, when a system of n similar molecules, at a given temperature T, is present in a large enough

volume v_0 so that it assumes the properties of a nondegenerate perfect gas, the corresponding sum-over-states can be expressed in the known form[9]

$$Z_{n,v_0,T}=\frac{1}{n!}\left(\sum_i \exp\left[-\epsilon_i(v_0)/kT\right]\right)^n, \quad (3.1)$$

where $\epsilon_i(v_0)$ is an eigenvalue of energy for a single molecule of the kind under consideration in the volume v_0, and we sum over all energy eigenstates i. In the second place, when the system is large enough so that it can be regarded as having a well-defined macroscopic pressure p, the correlation of thermodynamic with statistical-mechanical quantities provided by representing the thermodynamic equilibrium of a "closed" system by a canonical ensemble, shows that the change in sum-over-states with volume, holding composition and temperature constant, can be written in the form[10]

$$\frac{\partial \log Z}{\partial v}=\frac{p}{kT}. \quad (3.2)$$

Combining these two expressions, we may now write

$$\log Z_{n,v,T}=\log\frac{1}{n!}\left(\sum_i \exp\left[-\frac{\epsilon_i(v_0)}{kT}\right]\right)^n + \frac{1}{kT}\int_{v_0}^{v} p\,dv \quad (3.3)$$

as an expression for the sum-over-states corresponding to a system of n similar molecules at any desired volume v, and temperature T, where v_0 is a large enough volume so that the system then has the properties of a nondegenerate perfect gas.

In applying this expression to the problem outlined in the preceding section, we shall be interested in the value for the sum-over-states of a reservoir when the number of molecules therein is regarded as resulting from a fluctuation Δn away from the mean number of molecules $\bar{n}$ which the reservoir contains at equilibrium. Setting up expressions of the form (3.3) for the two cases when a system, at a given volume v and temperature T, contains $\bar{n}$ and $\bar{n}+\Delta n$ molecules, and subtracting one from the other, we readily obtain

$$\log\frac{Z_{\bar{n}+\Delta n}}{Z_{\bar{n}}}=\log\frac{\bar{n}!}{(\bar{n}+\Delta n)!}+\log\left(\sum_i \exp\left[-\frac{\epsilon_i(v_0)}{kT}\right]\right)^{\Delta n}+\frac{1}{kT}\int_{v_0}^{v} p_{\bar{n}+\Delta n}dv-\frac{1}{kT}\int_{v_0}^{v} p_{\bar{n}}dv \quad (3.4)$$

as an expression for the ratio of the sum-over-states $Z_{\bar{n}+\Delta n}$ for a system of $\bar{n}+\Delta n$ molecules at a given volume and temperature to the sum-over-states $Z_{\bar{n}}$ for the same system when it contains $\bar{n}$ molecules, where the volume v_0 appearing in the last three terms must be chosen large enough so that the system would behave as a nondegenerate perfect gas when it contains either $\bar{n}+\Delta n$ or $\bar{n}$ molecules in that volume, and where the symbols $p_{\bar{n}+\Delta n}$ and $p_{\bar{n}}$ in the last two terms denote the pressure of the system when it contains $\bar{n}+\Delta n$ or $\bar{n}$ molecules, respectively.

We must now discuss the dependence of the right-hand side of (3.4) on the value of the fluctuation Δn away from the mean molecular content $\bar{n}$, and on the value of the ratio of those two quantities $\Delta n/\bar{n}$. We shall be specially interested in the form of the right-hand side when the ratio $\Delta n/\bar{n}$ approaches zero, and shall wish to carry out the discussion in such a manner as to include not only the possibility of making this ratio small—with any given mean molecular content $\bar{n}$—by restricting attention to cases where the fluctuations Δn are sufficiently small, but also the possibility of making this ratio small—with any given value for the fluctuation Δn—by changing to a larger system (reservoir) with its mean molecular content $\bar{n}$ and its volume v increased in the same ratio.

To examine the dependence of the first term on the right-hand side of (3.4) on the quantities of interest we may rewrite it in the forms

[9] See for example Tolman, reference 2, Eq. (134.10).
[10] See for example Tolman, reference 2, Eq. (133.3).

$$\log\frac{\bar{n}!}{(\bar{n}+\Delta n)!}=\log\frac{1}{(\bar{n}+1)(\bar{n}+2)\cdots(\bar{n}+\Delta n)}$$

$$=-\log\bar{n}^{\Delta n}-\log\left(1+\frac{1}{\bar{n}}\right)\left(1+\frac{2}{\bar{n}}\right)\cdots\left(1+\frac{\Delta n}{\bar{n}}\right)$$

$$=-\Delta n\log\bar{n}-\left[\frac{1}{\bar{n}}-\frac{1}{2}\left(\frac{1}{\bar{n}}\right)^2+\frac{1}{3}\left(\frac{1}{\bar{n}}\right)^3-\cdots\right]-\left[\frac{2}{\bar{n}}-\frac{1}{2}\left(\frac{2}{\bar{n}}\right)^2+\frac{1}{3}\left(\frac{2}{\bar{n}}\right)^3-\cdots\right]$$

$$-\cdots-\left[\frac{\Delta n}{\bar{n}}-\frac{1}{2}\left(\frac{\Delta n}{\bar{n}}\right)^2+\frac{1}{3}\left(\frac{\Delta n}{\bar{n}}\right)^3-\cdots\right]. \quad (3.5)$$

As $\Delta n/\bar{n}$ goes to zero, either because we restrict our attention to cases where Δn is itself sufficiently small, or because we consider a large enough system to make $\bar{n}$ sufficiently large, this expression goes to

$$\log\frac{\bar{n}!}{(\bar{n}+\Delta n)!}=-\Delta n\log\bar{n}. \quad (3.6)$$

The explicit formulation of (3.5) is made for the case Δn positive, but a similar formulation can be made for the case Δn negative, which leads to the same limiting result (3.6).

The second term on the right-hand side of (3.4) is already expressed in a form which indicates its dependence on the quantities of interest, since it is seen to be the logarithm of a constant raised to the power Δn. It will be appreciated, however, that the value of this constant would be subject to alteration if we changed to a larger reservoir with a larger number of molecules, since we should then in general have to choose a larger volume v_0 to secure the behavior of a nondegenerate perfect gas.

The last two terms on the right-hand side of (3.4) may be re-expressed, in a form to show their dependence on the quantities of interest, by considering a Taylor's expansion for the value of the pressure of the system p as a function of its molecular content n. In doing this we shall assume that the substance in the system obeys an equation of state so that its pressure at a given temperature may be taken as a function of its density. This will let us put

$$p(n,v)=p(n/v)=p(\rho), \quad (3.7)$$

where we introduce the molecular density

$$\rho=n/v \quad \text{with} \quad \partial\rho/\partial n=1/v. \quad (3.8)$$

With the help of (3.7) and (3.8), we may now re-express the last two terms on the right-hand side of (3.4) in the forms

$$\frac{1}{kT}\int_{v_0}^{v}p_{\bar{n}+\Delta n}dv-\frac{1}{kT}\int_{v_0}^{v}p_{\bar{n}}dv=\frac{1}{kT}\int_{v_0}^{v}\left[\frac{1}{v}\frac{\partial p}{\partial\rho}\Delta n+\frac{1}{v^2}\frac{\partial^2 p}{\partial\rho^2}\frac{(\Delta n)^2}{2!}+\frac{1}{v^3}\frac{\partial^3 p}{\partial\rho^3}\frac{(\Delta n)^3}{3!}+\cdots\right]dv$$

$$=-\frac{\Delta n}{kT}\int_{\rho_0}^{\rho}\left[\frac{1}{\rho}\frac{\partial p}{\partial\rho}+\frac{1}{2}\frac{\partial^2 p}{\partial\rho^2}\frac{\Delta n}{\bar{n}}+\frac{1}{6}\rho\frac{\partial^3 p}{\partial\rho^3}\left(\frac{\Delta n}{\bar{n}}\right)^2+\cdots\right]d\rho, \quad (3.9)$$

where the derivatives of p with respect to ρ are to be taken when the system has the molecular content $n=\bar{n}$, and where the final form of expression has been obtained with the help of the substitutions $\bar{n}/v=\rho$ and $dv/v=-d\rho/\rho$. The final form of expression in (3.9) has been chosen, in a form depending on integration with respect to the density ρ, in order to make clear the consequences of changing to a larger system (reservoir) with the same proportional increase in molecular content $\bar{n}$

and in volume v. If such a change were made, it will be seen that the lower limit of integration ρ_0 would not need alteration since the properties of the substance could be taken as approaching those of a non-degenerate perfect gas at that given density, and the upper limit of integration and the functions of ρ appearing in the integrand would obviously not be altered. Hence as $\Delta n/\bar{n}$ goes to zero, either because we restrict our attention to cases where Δn is itself sufficiently small, or because we consider a large enough system to make $\bar{n}$ sufficiently large, we see, in accordance with the finite values to be expected for the derivatives of p with respect to ρ that occur in (3.9), that the last two terms on the right-hand side of (3.4) can be taken as going to

$$\frac{1}{kT}\int_{v_0}^{v} p_{\bar{n}+\Delta n}dv-\frac{1}{kT}\int_{v_0}^{v} p_{\bar{n}}dv=-\frac{\Delta n}{kT}\int_{\rho_0}^{\rho}\frac{1}{\rho}\frac{\partial p}{\partial \rho}d\rho. \tag{3.10}$$

Substituting (3.6) and (3.10) in (3.4), we can now write

$$\log\frac{Z_{\bar{n}+\Delta n}}{Z_{\bar{n}}}=-\Delta n\left(\log\bar{n}-\log\sum_i \exp\left[-\frac{\epsilon_i(v_0)}{kT}\right]+\frac{1}{kT}\int_{\rho_0}^{\rho}\frac{1}{\rho}\frac{\partial p}{\partial \rho}d\rho\right), \tag{3.11}$$

or

$$Z_{\bar{n}+\Delta n}=AB^{-\Delta n}, \tag{3.12}$$

where A and B are constants, as expressing in general, for a system of similar molecules at a given volume and temperature, the dependence of sum-over-states on fluctuations Δn in molecular content which are small compared with a particular value $\bar{n}$ for the number of molecules in the system.

§4. Distribution in Terms of Composition of System Proper

We are now ready to return to our previous expression (2.7) for the probability distribution for states of the system proper S, in terms of expressions for the sum-over-states of each of the reservoirs R_1, R_2, $\cdots R_h$. As a consequence of (3.12), we can now write

$$Z_{n_i'}=A_iB_i^{\bar{n}_i'-n_i'} \tag{4.1}$$

where A_i and B_i are constants, as a correct form of expression for the sum-over-states of the ith reservoir R_i, when it contains any number of molecules n_i' which is near enough the mean number $\bar{n}_1'$ so that the ratio $(n_i'-\bar{n}_i')/\bar{n}_i'$ is small. And by substituting the expression for n_i' given by (2.5) this can be rewritten in the form

$$Z_{n_i'}=A_iB_i^{\bar{n}_i'-n_i^0+n_i}, \tag{4.2}$$

where n_i^0 is the total number of molecules of the kind i in the combined system and n_i is the number of that kind in the system proper S.

Substituting expressions of the form (4.2) into our previous expression (2.7) for the probabilities of states of the system proper, we obtain

$$P_{n_1,n_2,\cdots n_h,s}=e^{(\psi-E_s)/\theta}\prod_{i=1}^{h}A_iB_i^{\bar{n}_i'-n_i^0+n_i}. \tag{4.3}$$

And by introducing new symbols defined by

$$e^{\Omega/\theta}=e^{\psi/\theta}\prod_{i=1}^{h}A_iB_i^{\bar{n}_i'-n_i^0} \quad\text{and}\quad e^{\mu_in_i/\theta}=B_i^{n_i}, \tag{4.4}$$

where the quantities Ω and μ_i are constants, we can then finally write

$$P_{n_1,n_2,\cdots n_h,s}=e^{(\Omega+\mu_1n_1+\mu_2n_2+\cdots+\mu_hn_h-E_s)/\theta}, \tag{4.5}$$

as the desired expression for the probability of any state s of the system proper, as determined by its molecular contents n_1, n_2, $\cdots n_h$, and energy E_s, when in that state.

It will be noted that the above dependence on composition and energy is that for a grand canonical distribution. It will be seen, in accordance with the derivation of (4.5), that this distribution can be taken as valid, in the neighborhood of the mean molecular composition of the system proper, for any fluctuation Δn_i in the number of molecules of any kind present therein, which can be regarded as vanishingly small compared with the mean number n_i' present in the corresponding reservoir. It will also be seen, moreover, that the range of validity can be made as wide as desired by taking large enough reservoirs with large enough mean molecular contents $\bar{n}_i'$. Since the change to a larger reservoir may be made by taking the same proportional

increase in volume and in mean molecular content, it will be appreciated that such a change need involve no major alteration in the escaping tendency for the molecules in the reservoir or in the character of the equilibrium in the system proper.

The foregoing derivation has been carried out for simplicity for the case when the system proper S is taken as connected through a semi-permeable membrane with a separate reservoir for each of the different kinds of molecules involved. Similar derivations can also be carried out, however, for cases when the system is taken as connected through suitable membranes with reservoirs for more than a single substance, or indeed as connected—without any membrane being necessary—with a single reservoir for all the component substances. In treating such cases, the sum-over-states for a reservoir, containing more than a single kind of molecule, can now be obtained by starting with the known expression for sum-over-states when a mixture of molecules is present in a large enough volume to act as a nondegenerate perfect gas, and then adding as before the increase in sum-over-states that occurs on compression to the volume of interest. The dependence of the result on molecular constitution can then be investigated, much as before, the main difference lying in the necessity of now introducing a Taylor's expansion, which gives the pressure of the reservoir contents as a function of changes in the numbers of more than a single kind of molecules. The range of validity for the grand canonical distribution is still found in such cases to be limited only by the restriction to fluctuations, around the mean number of molecules of any constituent in the system proper, which are vanishingly small compared with the mean number in the appropriate reservoir. Hence, under such circumstances, we may now take the grand canonical distribution as applying in general to the equilibrium condition for a system proper which is connected with one or more suitable reservoirs for providing its constituent substances.

§5. Concluding Remarks

We may now make some concluding remarks, as to the conditions leading to the establishment of grand canonical distributions, and as to the propriety of using grand canonical ensembles for representing equilibrium in the case of "open" thermodynamic systems.

In accordance with the previously known results of statistical mechanics, we can represent the establishment of equilibrium in a "closed" system of molecules, left in thermal contact with a large heat bath at temperature T, by the establishment of a canonical distribution with $\theta=kT$ in the corresponding representative ensemble, and can then use the canonical ensemble thus arising to represent the condition of equilibrium in the "closed" system of interest. In accordance with the results of the present article, we have found that a canonical distribution in the representative ensemble for a "closed" system, consisting of a system proper connected with a suitable reservoir or reservoirs for furnishing the various kinds of molecules involved, implies a grand canonical distribution, in that part of the ensemble which represents the system proper, for all fluctuations in molecular contents which are vanishingly small compared with the mean contents available in the reservoirs. Hence, introducing the usual term "open" system to denote a system proper which is connected with one or more reservoirs of large enough capacity for the satisfactory accommodation of all fluctuations of interest, we can now represent the establishment of equilibrium, in an "open" system of molecules at temperature T, by the establishment of a grand canonical distribution in the corresponding representative ensemble with $\theta=kT$, and can then use the grand canonical ensemble thus arising to represent the condition of equilibrium in the "open" system under consideration.

In making such use of grand canonical ensembles to represent the condition of equilibrium, in actual situations where a system of interest is connected with one or more reservoirs, cases may arise where the range of validity for the grand canonical distribution is not wide enough to cover all fluctuations in the composition of the system which are of interest. This may occur either because the reservoirs are so small as to make the actual range of validity a very narrow one, or because the character of the system is such as to give a high probability for compositions of the system differing widely from the

mean,[11] thus making the range of interest over which we desire accuracy a wide one. In such cases, we have the possibility of giving as accurate a treatment of the probabilities for different compositions of the system as may be necessary, by including higher order terms of the kind $\Delta n/\bar{n}$ in equations such as (3.5) and (3.9). It is to be emphasized, however, in the usual situations where we should wish to apply the grand canonical distribution, that the typical cases which arise are those where the numbers of molecules available in the reservoirs are great enough, and the probabilities for compositions of the system widely different from the mean are small enough, so that the grand canonical ensemble gives an appropriate description of equilibrium.

It should be remarked that the circumstances, which lead to the grand canonical rather than the canonical distribution as the appropriate description of equilibrium, are perhaps more frequently present in nature than is usually appreciated, owing to the frequency with which we are interested in the behavior of a system of molecules which in actuality forms part of, or has been cut off, from a larger system. From a mathematical point of view, this is a fortunate happening, since, in spite of its apparently greater complexity of form, the grand canonical distribution really proves easier to handle than the canonical distribution, owing to the fact that summations over different possible states can be carried out without any limitation on the total number of molecules. Thus in the case of degenerate Einstein-Bose and Fermi-Dirac gases it is found, when the interaction between molecules can be neglected, that the usual expressions for the mean numbers of molecules in different molecular states can be simply and accurately derived for a grand canonical ensemble, although holding only approximately in a canonical ensemble.[12]

In conclusion, it may be emphasized that the foregoing demonstration of an actual tendency toward the establishment of grand canonical distributions in the ensembles by which we represent the establishment of equilibrium in "open" systems, is of assistance in placing on a sound and understandable basis the use which has been made of grand canonical ensembles for the representation of thermodynamic equilibrium in "open" systems. Previous work[7] has shown that this use of grand canonical ensembles provides consistent statistical-mechanical analogs for thermodynamic quantities, in particular analogs for the Gibbs' potentials which give a statistical-mechanical explanation of the known thermodynamic conditions for equilibrium towards the transfer of component substances between systems placed in connection, and an analog for entropy which gives a statistical-mechanical explanation for the thermodynamic principle of the precise additivity of entropies when similar homogeneous systems are combined thus solving the so-called Gibbs paradox. It has not hitherto been apparent, however, that the introduction of grand canonical ensembles to represent the equilibrium of "open" thermodynamic systems is not only suggested by the consistency of the thermodynamic analogies thus provided, but is also dictated by the actual tendency for grand canonical distributions to be established in the ensembles by which we must represent the establishment of equilibrium in "open" systems, if we apply the accepted principles of statistical mechanics.

[11] A judgment as to the probabilities for compositions differing from the mean can be obtained with the help of the formula for the mean square fluctuations in composition in a grand canonical ensemble, see Gibbs, reference 2, Eq. (541); Tolman, reference 2, Eq. (141.42).

[12] See Pauli, Zeits. f. Physik **41**, 81 (1927); Tolman, reference 2, §114.

A New Method for Calculating Wave Functions in Crystals

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(Received April 8, 1940)

For many problems in the electron theory of metals none of the methods hitherto used to calculate the eigenfunctions and energy values of an electron in a crystal lattice is satisfactory. It is here proposed that these wave functions and energies be calculated by solving a secular equation with wave functions χ_k which are simply plane waves made orthogonal to the core eigenfunctions. The rapidity of convergence to be expected for such a procedure is discussed. Some methods for practical computation are suggested, and expressions are given for the matrix elements occurring in the secular equation.

1. INTRODUCTION

THE problem of calculating explicitly the eigenfunctions and energy values for an electron in the periodic field of a crystal is one which has not yet been satisfactorily solved. The method of Wigner and Seitz[1] is simple and gives good results for the lower states of the valence electron band of a metal, but the natural extension of this method to states of higher energy[2] becomes rapidly more unreliable as the energy increases.[3] Another method, proposed by Slater,[4] is better adapted to high energy states, but has the disadvantage that the fluctuations in potential in the outer region of each atomic cell are disregarded. The method to be described in the present paper, while not entirely satisfactory, possesses advantages which make it applicable to numbers of problems which cannot be treated safely by the other methods. The principal disadvantage of the procedure to be described is that it is rather laborious. It is nevertheless practicable, and some successful applications to simple metals have already been made.[5]

One of the earliest methods of approximating to the electron energies in a crystal consisted in starting from plane waves and treating the periodic variation of potential as a perturbation.[6] This method has been considered futile for accurate work, however. For if we set up the matrix of the Hamiltonian relative to some number n of plane waves, and solve the secular equation, we find that for small values of n the lowest root approximates a valence electron eigenvalue, but that as $n\rightarrow\infty$ the lowest root approaches the energy of the lowest state of an atomic core, and it is only one of the higher roots which approaches a valence electron eigenvalue.[7] And to get a good final approximation to the valence electron energy, we would have to use a very large value of n, so that the solution of the secular equation would be prohibitively difficult. This corresponds to the fact that in the Fourier expansion of a valence electron eigenfunction there is a great contribution from waves of short wave-length. However, if we set up instead the matrix of the Hamiltonian relative to a set of n functions which are all orthogonal to the eigenfunctions of the core bands of the crystal, and solve the secular equation for this matrix, the lowest root must approach a valence electron eigenvalue as $n\rightarrow\infty$, and it is more reasonable to expect that the convergence will be rapid.

Two principles are involved in the procedure to be described. The first is that the statement at the end of the preceding paragraph is approximately true if we work with wave functions which are orthogonal, not to the true core eigenfunctions φ_{jk}, but to functions φ_{jk}' which closely approximate the φ_{jk}. Let χ_k be the wave function obtained by orthogonalizing a plane wave of wave vector $\mathbf{k}$ to all the functions φ_{jk}'

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[1] E. Wigner and F. Seitz, Phys. Rev. **43**, 804 (1933); **46**, 509 (1934).
[2] J. C. Slater, Phys. Rev. **45**, 794 (1934).
[3] W. Shockley, Phys. Rev. **52**, 866 (1937).
[4] J. C. Slater, Phys. Rev. **51**, 846 (1937).
[5] C. Herring and A. G. Hill, Phys. Rev., to appear later.
[6] See the article of Sommerfeld and Bethe, *Handbuch der Physik* (2nd ed., Berlin, 1933), Vol. 24/2, p. 385.
[7] Cf. J. C. Slater, Rev. Mod. Phys. **6**, 250–258 (1934).

having the same reduced wave vector.[8] Let φ_k' be the wave function of lowest average energy which can be constructed from the functions $\chi_{k+2\pi g}$, where $\mathbf{k}$ is a given vector and $\mathbf{g}$ runs over all vectors of the reciprocal lattice of the crystal. The average energy E_k' of φ_k' is of course the limit, as $n\to\infty$, of the smallest root of the secular equation constructed with the n functions χ of smallest wave vectors. This E_k' is slightly less than the eigenvalue E_k of the lowest valence electron state with the same reduced wave vector.

If we use the abbreviation

$$(\varphi_{sk}, \varphi'_k)=\int \varphi_{sk}^*\varphi_k' d\tau$$

we may write

$$\varphi_k'=\sum_s (\varphi_{sk}, \varphi_k')\varphi_{sk},$$
$$E_k'=\sum_s |(\varphi_{sk}, \varphi_k')|^2 E_{sk},$$

where the φ_{sk} and E_{sk} are the true eigenfunctions and energies of all the core and valence electron states with reduced wave vector $\mathbf{k}$. Since φ_k' is orthogonal to any φ_{jk}' we have by Bessel's inequality

$$|(\varphi_{sk}, \varphi_k')|^2+|(\varphi_{sk}, \varphi_{jk}')|^2\leqslant 1,$$

so

$$E_k-E_k'=\sum_s |(\varphi_{sk}, \varphi_k')|^2(E_k-E_{sk})\leqslant \sum_{j\,\text{core}} |(\varphi_{jk}, \varphi_k')|^2(E_k-E_{jk})\leqslant \sum_{j\,\text{core}} [1-|(\varphi_{jk}, \varphi_{jk}')|^2](E_k-E_{jk}). \quad (1)$$

This tells us how much we can allow φ_{jk}' to deviate from the corresponding true eigenfunction φ_{jk} of the core band, when an allowable error (E_k-E_k') has been specified. Moreover, φ_k can be obtained from φ_k' by adding multiples of the true core functions and renormalizing. So if the right of (1) is sufficiently small, it is reasonable to expect that we can get a much better approximation to φ_k, and hence to E_k, by adding to φ_k' a small multiple of each φ_{jk}'; i.e., we can treat the matrix elements of the Hamiltonian connecting φ_k' with the φ_{jk}' as small perturbations.

The second of the principles mentioned has to do with the fact that in the troublesome region near a nucleus, the solutions of the wave equation do not depend very much on the energy, because the negative potential is very large. Thus for example the s part of a valence electron eigenfunction φ_k can be approximated near a nucleus by an s solution of the wave equation for a somewhat different energy, the p part by a p solution for a different energy, etc. So by subtracting off from φ_k a suitable multiple of some s solution of the wave equation we can obtain a function which no longer has a sharp maximum at the nucleus, and which accordingly can be much better represented by a few terms of its Fourier series than could the original φ_k.

This suggests that it would be practical to try to approximate φ_k by a linear combination of a few plane waves, plus a linear combination of a few functions u_j localized about each nucleus and obeying wave equations of the form

$$\nabla^2 u_j+(E_j-V_j)u_j=0 \quad (2)$$

(distances to be measured in Bohr units and energies in rydbergs), where E_j and V_j are such that within a reasonable radius from the nucleus

$$(E_j-V_j)-(E_k-U)\ll(E_k-U),$$

U being the actual potential in the crystal. By considering the case where V_j is nearly the same as U and the u_j are atomic core functions it is possible to show that the lowest root of the secular equation mentioned in the preceding paragraphs ought to converge fairly rapidly toward E_k'. For definiteness let us suppose V_j to be spherically symmetrical, and the u_j to be $1s$, $2s$, and $2p$ functions. Then near the nucleus we can represent the s part of φ_k fairly well by a linear combination of the $1s$ and $2s$ functions and the p part by a multiple of the $2p$ function. The d part of φ_k will vary rather smoothly with position near the nucleus, if the atomic number is reasonably small. Thus we can choose coefficients $\alpha_{j\nu}$ so that

$$\varphi_k-\sum_{j\nu} \alpha_{j\nu}u_j(\mathbf{r}-\mathbf{R}_\nu) \quad (3)$$

[8] For the conception of "reduced wave vector" see Sommerfeld and Bethe, reference 6, p. 394.

varies much more gradually near each nuclear position than does φ_k. The Fourier series for the function (3) will then converge rapidly, and so φ_k can be fairly well approximated by a linear combination of a few plane waves plus the sum $\sum_{j\nu}\alpha_{j\nu}u_j(\mathbf{r}-\mathbf{R}_\nu)$. Now if the V_j resemble U sufficiently closely, the Bloch waves[9] constructed from the $u_j(\mathbf{r}-\mathbf{R}_\nu)$ will be good approximations to the core eigenfunctions φ_{jk}, and can be used for the φ_{jk}' mentioned in the preceding paragraph. In such case let us write

$$\varphi_k=\sum_{j\nu}\alpha_{j\nu}u_j(\mathbf{r}-\mathbf{R}_\nu)+\sum_{\mathbf{g}}\frac{\beta_\mathbf{g}}{\Omega^{\frac{1}{2}}}\exp i(\mathbf{k}+2\pi\mathbf{g})\cdot\mathbf{r},$$

where Ω is the volume of the crystal. Then from the fact that φ_k is approximately orthogonal to each u_j we may conclude that

$$\varphi_k\approx\sum_{\mathbf{g}}\beta_\mathbf{g}\chi_{k+2\pi\mathbf{g}},$$

where each χ is simply a plane wave made orthogonal to all the $u_j(\mathbf{r}-\mathbf{R}_\nu)$. From what has been said above this series will converge rapidly. It is thus to be expected that the lowest root of the secular equation formed using the first n χ's will converge rapidly toward E_k' as n is increased.

In many cases it will be sufficient to work with a secular equation formed from the χ's, and Sections 2 and 3 will be concerned exclusively with such cases. It should be mentioned, however, that it may sometimes be desirable to use one or more functions u_i which do not resemble core functions, and to set up a secular equation using these u_i together with χ-functions which are orthogonal to the other u_j. Consider for example a crystal (such as Li or Be) for which there is only one core band, derived from the 1s atomic levels. First assume the u_{1s} to be the 1s eigenfunction in a central field V which is the spherical average of the true field U. If near a nucleus

$$V(r)=-2Z/r+a+br\cdots,$$

we have

$$u_{1s}\propto 1-Zr+\left(\frac{Z^2}{3}+\frac{a-E_{1s}}{6}\right)r^2+\cdots,$$

while, to a high degree of accuracy,

$$s\text{ part of }\varphi_k\propto 1-Zr+\left(\frac{Z^2}{3}+\frac{a-E_k}{6}\right)r^2+\cdots.$$

Thus with properly chosen coefficients the s part of the function (3) will vanish with its radial derivative at each nucleus, and even the second radial derivative will be small. The p part of φ_k, on the other hand, has quite a large gradient at a nucleus, whereas the p part of a sum of a few χ-functions (which is the same as the p part of a sum of plane waves) cannot have a very large gradient unless waves of short wave-length are included.[10] Thus if we use only a few χ-functions we can approximate the s part of φ_k much more closely than the p part. The situation could be improved by inventing a p function u_p obeying near the nucleus a wave equation resembling that for the p part of φ_k, and dying off smoothly to zero in the outer regions of an atomic cell. A combination of a few χ-functions (or plane waves) with a suitable multiple of u_p should then give a much closer approximation to the p part of φ_k. Our final wave function need not be even approximately orthogonal to u_p, however, since there is no p band in the core.

2. General Theory

Consider a crystal of volume Ω, containing N identical unit cells each of volume Ω_0. For simplicity assume that all atoms of the crystal are of the same kind, even though there may be more than one atom per unit cell. Let $U(\mathbf{r})$ be the potential energy of an electron at the point $\mathbf{r}$. Let $V(r)$ be a more or less arbitrary spherically symmetric potential field, chosen, however, so as to resemble U as closely as possible in the region near a nucleus. When calculations are made for several values of the lattice constant or of other parameters, U will be a different function in each case, whereas V is best kept fixed to avoid repetition of the labor of computing core functions and quantities which depend on them. Let one or more real functions u_{nlm} be core eigenfunctions in the field $V(r)$, with

[9] See Sommerfeld and Bethe, reference 6, p. 394.
[10] Cf. Fig. 4 of the following paper.

energies E_{nlm}. The Bloch waves formed from these "atomic" functions will play the part of the functions which were called $\varphi_{jk'}$ in the preceding section.

In the following it will be assumed that energies are measured in rydbergs and distances in units of the Bohr radius. The functions χ_k we shall use will be defined by

$$N^{\frac{1}{2}}\chi_k(\mathbf{r})=\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{(\Omega_0)^{\frac{1}{2}}}-\sum_\nu\sum_{nl}\exp(i\mathbf{k}\cdot\mathbf{R}_\nu)A_{nl}(k)u_{nl;\,k}(\mathbf{r}-\mathbf{R}_\nu), \tag{4}$$

where ν runs over all atoms of the crystal, $u_{nl;\,k}$ is the function u_{nl0} when the axis to which m refers is chosen in the direction of $\mathbf{k}$, and where

$$A_{nl}(k)=\int_\infty\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{(\Omega_0)^{\frac{1}{2}}}u_{nl;\,k}d\tau. \tag{5}$$

It is easily verified that each χ_k is orthogonal to all the $u_{nlm}(\mathbf{r}-\mathbf{R}_\nu)$ if one neglects the fact that core functions about different atoms may overlap and thus fail to be exactly orthogonal to one another.

Before writing down the accurate expressions for the matrix elements of the Hamiltonian between the functions (4), which are rather lengthy, it may be worth while to see what they look like if one makes the approximations that the $u_{nlm}(\mathbf{r}-\mathbf{R}_\nu)$ are true eigenfunctions in the crystal field and that the functions on different atoms do not overlap. Noting that

$$\int_\infty u_{nl;\,k}(\mathbf{r})u_{nl;\,k'}(\mathbf{r})d\tau=P_l(\cos\theta_{kk'}),$$

where $\theta_{kk'}$ is the angle between $\mathbf{k}$ and $\mathbf{k}'$, we have in the approximation mentioned

$$(\chi_k,\,\chi_{k'})=\delta_{kk'}-\sum_{nl}A_{nl}(k)A_{nl}(k')P_l(\cos\theta_{kk'})\sum_\nu^{\text{cell}}\exp[i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_\nu], \tag{6}$$

if $(\mathbf{k}-\mathbf{k}')$ is 2π times a vector of the reciprocal lattice, and $=0$ otherwise. The summation in (6) is over all atoms ν in a single unit cell. Also, since in our present approximation the Hamiltonian H applied to each $u_{nl;\,k}$ merely multiples it by E_{nl},

$$(\chi_k,\,H\chi_{k'})=k^2\delta_{kk'}+U[\mathbf{k}'-\mathbf{k}]-\sum_{nl}E_{nl}A_{nl}(k)A_{nl}(k')P_l(\cos\theta_{kk'})\sum_\nu^{\text{cell}}\exp[i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_\nu], \tag{7}$$

if $(\mathbf{k}'-\mathbf{k})$ is 2π times a vector of the reciprocal lattice, and $=0$ otherwise. Here

$$U[\mathbf{K}]=\frac{1}{\Omega_0}\int_{\Omega_0}\exp(i\mathbf{K}\cdot\mathbf{r})\,U(\mathbf{r})d\tau \tag{8}$$

is a Fourier coefficient of the potential.

Numerical calculations have shown that solution of a secular equation obtained from a few χ-functions from (6) and (7) gives a fair approximation to the energy of a valence electron eigenfunction for a metal, such as an alkali, for which the u_{nlm} are confined quite closely around the nuclei. However, the derivatives of energy with respect to lattice constant, as obtained from (6) and (7), approximate the true derivatives very poorly. This is not surprising, for a small change in lattice constant has a large effect on the validity of the assumption that U and V are identical, and of the assumption that core functions on different atoms do not overlap. We therefore need exact expressions for $(\chi_k,\,\chi_{k'})$ and $(\chi_k,\,H_{k'})$. When $(k'-k)$ is 2π times a vector of the reciprocal

lattice, we have

$$(\chi_k, H\chi_{k'}) = k^2\delta_{kk'} + U[\mathbf{k}'-\mathbf{k}] - \frac{1}{N}\sum_{\nu'}\sum_{nl}\int_\Omega \Big[\frac{\exp(-i\mathbf{k}\cdot\mathbf{r})}{(\Omega_0)^{\frac{1}{2}}}\exp(i\mathbf{k}'\cdot\mathbf{R}_{\nu'})A_{nl}(k')Hu_{nl;\,k'}(\mathbf{r}-\mathbf{R}_{\nu'})$$

$$+\frac{\exp(i\mathbf{k}'\cdot\mathbf{r})}{(\Omega_0)^{\frac{1}{2}}}\exp(-i\mathbf{k}\cdot\mathbf{R}_{\nu'})A_{nl}(k)Hu_{nl;\,k}(\mathbf{r}-\mathbf{R}_{\nu'})\Big]d\tau$$

$$+\frac{1}{N}\sum_{\nu,\nu'}\sum_{nl,n'l'}\int_\Omega \exp(-i\mathbf{k}\cdot\mathbf{R}_\nu + i\mathbf{k}'\cdot\mathbf{R}_{\nu'})A_{nl}(k)A_{n'l'}(k')u_{nl;\,k}(\mathbf{r}-\mathbf{R}_\nu)Hu_{n'l';\,k'}(\mathbf{r}-\mathbf{R}_{\nu'})d\tau. \quad (9)$$

In (9) we may put $H=[-\nabla^2+V(\mathbf{r}-\mathbf{R}_{\nu'})]+[U(\mathbf{r})-V(\mathbf{r}-\mathbf{R}_{\nu'})]$. To simplify the resulting expression, three new symbols will be introduced:

$$I_{nl}(\mathbf{k};\mathbf{k}';\nu) = \int_\infty \frac{\exp[i\mathbf{k}\cdot(\mathbf{r}-\mathbf{R}_\nu)]}{(\Omega_0)^{\frac{1}{2}}}[U(\mathbf{r})-V(\mathbf{r}-\mathbf{R}_\nu)]u_{nl;\,k'}(\mathbf{r}-\mathbf{R}_\nu)d\tau, \quad (10)$$

$$S(nl\mathbf{k}\nu, n'l'\mathbf{k}'\nu') = \int_\infty u_{nl;\,k}(\mathbf{r}-\mathbf{R}_\nu)u_{n'l';\,k'}(\mathbf{r}-\mathbf{R}_{\nu'})d\tau, \quad (11)$$

$$F(nl\mathbf{k}\nu, n'l'\mathbf{k}'\nu') = \int_\infty u_{nl;\,\mathbf{k}}(\mathbf{r}-\mathbf{R}_\nu)[U(\mathbf{r})-V(\mathbf{r}-\mathbf{R}_{\nu'})]u_{n'l';\,k'}(\mathbf{r}-\mathbf{R}_{\nu'})d\tau. \quad (12)$$

Inserting (10), (11), and (12) into (9) gives finally

$$(\chi_k, H\chi_{k'}) = k^2\delta_{kk'} + U[\mathbf{k}'-\mathbf{k}] - \sum_{nl}E_{nl}A_{nl}(k)A_{nl}(k')P_l(\cos\theta_{kk'})\sum_\nu^{\text{cell}}\exp[i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_\nu]$$

$$-\sum_{nl}\sum_\nu^{\text{cell}}[(A_{nl}(k')I_{nl}^*(\mathbf{k};\mathbf{k}';\nu)+A_{nl}(k)I_{nl}(\mathbf{k}';\mathbf{k};\nu)]\exp[i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_\nu]$$

$$+\sum_{nl,n'l'}A_{nl}(k)A_{n'l'}(k')\sum_\nu^{\text{cell}}F(nl\mathbf{k}\nu, n'l'\mathbf{k}'\nu)\exp[i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_\nu]$$

$$+\sum_{nl,n'l'}A_{nl}(k)A_{n'l'}(k')\sum_{\nu'\neq\nu}\sum_\nu^{\text{cell}}[E_{nl}S(nl\mathbf{k}\nu, n'l'\mathbf{k}'\nu')$$

$$+F(nl\mathbf{k}\nu, n'l'\mathbf{k}'\nu')]\exp[-i\mathbf{k}\cdot\mathbf{R}_\nu + i\mathbf{k}'\mathbf{R}_{\nu'}], \quad (13)$$

if $(\mathbf{k}-\mathbf{k}')$ is 2π times a vector of the reciprocal lattice, and $=0$ otherwise. Similarly

$$(\chi_k, \chi_{k'}) = \delta_{kk'} - \sum_{nl}A_{nl}(k)A_{nl}(k')P_l(\cos\theta_{kk'})\sum_\nu^{\text{cell}}\exp[i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_\nu]$$

$$+\sum_{nln'l'}A_{nl}(k)A_{n'l'}(k')\sum_{\nu'\neq\nu}\sum_\nu^{\text{cell}}S(nl\mathbf{k}\nu, n'l'\mathbf{k}'\nu')\exp(-i\mathbf{k}\cdot\mathbf{R}_\nu + i\mathbf{k}'\cdot\mathbf{R}_{\nu'}), \quad (14)$$

if $(\mathbf{k}'-\mathbf{k})$ is 2π times a vector of the reciprocal lattice, and $=0$ otherwise. When there are core functions with $l>0$, the S integrals and the F integrals for $\nu\neq\nu'$ become very difficult to evaluate, so in such cases it will be convenient to choose core functions which become zero at a value of r less than half the distance between neighboring atoms. If this is done the last terms of (13) and (14) disappear.

The matrix elements (13) and (14) determine the wave function $\varphi_k' = \sum_g a_g\chi_{k+2\pi g}$ mentioned in Section 1, whose average energy E_k' is less than, but nearly equal to, the eigenvalue E_k we are

seeking. If the correction bounded by (1) is sufficiently small, it will be a good approximation to set

$$E_k = E_k' + \sum_{nlm\nu} \frac{|[\varphi_k', \{H-E_k'\}u_{nlm}(\mathbf{r}-\mathbf{R}_\nu)]|^2}{E_k'-E_{nl}}. \tag{15}$$

For brevity only matrix elements of the form $[\chi_k, Hu_{nl;k'}(\mathbf{r}-\mathbf{R}_\nu)]$ will be given here; the summation on m in (15) is easily expressed in terms of these when $l=0$ or 1:

$$\exp(i\mathbf{k}'\cdot\mathbf{R}_\nu)N^{\frac{1}{2}}[\chi_{k'}, Hu_{nl;k}(\mathbf{r}-\mathbf{R}_\nu)] = I_{nl}^*(\mathbf{k}';\mathbf{k};\nu) - \sum_{n'l'} A_{n'l'}(k')F(n'l'\mathbf{k}'\nu, nl\mathbf{k}\nu)$$
$$- \sum_{n'l'} A_{n'l'}(k') \sum_{\nu'\neq\nu} [E_{nl}S(n'l'\mathbf{k}'\nu', nl\mathbf{k}\nu) + F(n'l'\mathbf{k}'\nu', nl\mathbf{k}\nu)] \exp[i\mathbf{k}'\cdot(\mathbf{R}_\nu-\mathbf{R}_{\nu'})] \tag{16}$$

$$\exp(i\mathbf{k}'\cdot\mathbf{R}_\nu)N^{\frac{1}{2}}[\chi_{k'}, u_{nl;k}(\mathbf{r}-\mathbf{R}_\nu)] = -\sum_{n'l'} A_{n'l'}(k) \sum_{\nu'\neq\nu} S(n'l'\mathbf{k}'\nu', nl\mathbf{k}\nu) \exp[i\mathbf{k}'\cdot(\mathbf{R}_\nu-\mathbf{R}_{\nu'})]. \tag{17}$$

When core functions on different atoms are so constructed that they do not overlap, (17) and the last term of (16) vanish.

3. Remarks and Discussion

(1) Choice of U and evaluation of $U[\mathbf{K}]$. In metallic crystals it is convenient and accurate to assume that U is a summation of potentials $v(\mathbf{r}-\mathbf{R}_\nu)$ due to the different ions, plus the potential due to a homogeneous free electron distribution. In such case $U[\mathbf{K}]$ can be reduced for $\mathbf{K}\neq 0$ to

$$U[\mathbf{K}] = \frac{3}{Kr_s^3}\left(\frac{1}{n}\sum_{\nu}^{\text{cell}} \exp(i\mathbf{K}\cdot\mathbf{R}_\nu)\right)\left(-\frac{2X}{K} - \int_0^\infty r\left[v(r)+\frac{2X}{r}\right]\sin Kr dr\right). \tag{18}$$

where n is the number of atoms in a unit cell and X is the number of valence electrons per atom, so that $v(r)\sim -2X/r$ for large r. The most convenient choice for the zero of U is that made by Wigner and Seitz: the average value of U over a unit cell is defined to equal the average over an s sphere of the potential due to the ion at its center and an amount of negative charge sufficient to neutralize it, uniformly distributed throughout the s sphere. Thus

$$U[0] = \frac{2.4}{r_s} + \frac{3}{r_s^3}\int_0^{r_s} r^2 v(r)dr, \tag{19}$$

provided $v(r) = -2X/r$ for $r \geqslant r_s$.

(2) Choice of V. The perturbation in (15) due to the core states can be made quite small by choosing a V which resembles U very closely; for the above choice of U we might use

$$V = v(r) + \frac{3X}{r_0} - \frac{Xr^2}{r_0^3} \quad \text{for } r \leqslant r_0, \tag{20}$$
$$= 0 \quad \text{for } r \geqslant r_0.$$

Thus for $r \leqslant r_0$, V coincides with the spherical average of U[11] when r_s has the value r_0. The disadvantage of such a choice is that the u_{nlm} extend to infinity, so that the overlap integrals (10), (11), and (12) are difficult to evaluate. When the only core functions are the $1s$, it is not hard to evaluate the S integrals exactly, and rough approximations can be obtained to the other integrals which are adequate for many purposes (see below). But when there are core functions with $l>0$, or when the s functions are too spread out for these rough values to suffice, the evaluation of the overlap

[11] This is strictly true only if the small correction of Appendix II, reference 1, is neglected.

integrals seems hopelessly difficult. In such cases it will probably be best to construct u_{nlm} functions which vanish at less than half the distance between neighboring atoms; this of course means that V must become infinite.

If we require $u_{nlm}(r_1, \theta, \varphi)=0$, it is desirable that $\partial u_{nlm}/\partial r$ vanish at $r=r_1$ also, since we want the Fourier expansion of (3) to converge rapidly. At the same time the right of (1) must be kept small. If $u_{nlm}{}^{(0)}$ are the eigenfunctions in the field (20), the Bloch waves constructed from them will resemble the true core eigenfunctions much more closely than those constructed from the present u_{nlm}. So remembering that $u_{nlm}{}^{(0)}$ and u_{nlm} are assumed normalized we have, for the average over the $(2l+1)$ bands arising from the atomic states of given n and l,

$$\mathrm{Av}[1-|(\varphi_{jk}, \varphi_{jk}')|^2]\approx 1-|(u_{nlm}{}^{(0)}, u_{nlm})|^2=\tfrac{1}{2}\int_\infty |u_{nlm}{}^{(0)}-u_{nlm}|^2 d\tau$$

$$=\tfrac{1}{2}\int_0^\infty (P_{nl}{}^{(0)}-P_{nl})^2 dr>\tfrac{1}{2}\int_{r_1}^\infty P_{nl}{}^{(0)2} dr, \quad (21)$$

where

$$u_{nlm}=\frac{P_{nlm}(r)\,Y_{lm}(\theta, \varphi)}{r}, \qquad \int_0^\infty P_{nl}{}^2 dr=1, \quad \text{etc.}$$

If P_{nl} is defined to be a multiple of $P_{nl}{}^{(0)}$ for $r\leqslant r_2$, and a simple analytic function of r (e.g., a polynomial) for $r_2\leqslant r\leqslant r_1$, so chosen that P_{nl} and P_{nl}' are continuous at r_2 and vanish at r_1, the left of (21) will usually come out of the order of 3 or 4 times the integral

$$\tfrac{1}{2}\int_{r_1}^\infty P_{nl}{}^{(0)2} dr;$$

from this and (1) one can estimate in advance whether or not (15) will suffice for the core perturbation.

(3) Evaluation of the S integrals. If the field (20) is used, we may write

$$P_{1s}(r)=(4\pi)^{\frac{1}{2}} r u_{1s}(r)=B\exp[-(-E)_{1s}{}^{\frac{1}{2}} r]-\gamma(r), \quad (22)$$

where $\gamma(r)$ vanishes for $r\geqslant r_0$. The average of $(1/r)\exp[-(-E)_{1s}{}^{\frac{1}{2}} r]$ over the surface of a sphere of radius ρ whose center is at a distance d from the origin is

$$\frac{\exp[-(-E)_{1s}{}^{\frac{1}{2}} d]}{d(-E_{1s})^{\frac{1}{2}}}\,\frac{\sinh(-E_{1s})^{\frac{1}{2}}\rho}{\rho}.$$

From this and by neglecting the overlapping of γ's on different atoms the S integral can be reduced to an expression containing a single radial integral:

$$S(\nu, \nu')\equiv S(1s\mathbf{k}\nu, 1s\mathbf{k}'\nu')=\frac{\exp[-(-E)_{1s}{}^{\frac{1}{2}}|\mathbf{R}_\nu-\mathbf{R}_{\nu'}|]}{2(-E)_{1s}{}^{\frac{1}{2}}}\left[B^2-\frac{2B}{|\mathbf{R}_\nu-\mathbf{R}_{\nu'}|}\int_0^\infty \gamma(r)\sinh[(-E)_{1s}{}^{\frac{1}{2}} r]dr\right]. \quad (23)$$

(4) The I_{nl} and F integrals. If the field (20) is used, and the field U is as described above, the contributions to I_{nl} and to the F integrals for $\nu=\nu'$ may be divided into two parts: that from inside a radius r_0 and that from outside r_0. The first of these is the more important when r_s differs appreciably from r_0. This suggests that we may approximate U by $v(r)+3X/r_s-Xr^2/r_s{}^3$, which is its spherical average[12] for $r\leqslant r_s$, and approximately its spherical average for somewhat greater values of r. This approximation should be very good indeed for the F integral with $\nu=\nu'$, less so for the I_{nl}.

[12] This is strictly true only if the small correction of Appendix II, reference 1, is neglected.

If this is inserted in (10) we have, for a $1s$ function

$$I_{1s}(\mathbf{k}) \equiv I_{1s}(\mathbf{k};\mathbf{k}';\nu) \approx \int_{\infty} \frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{(\Omega_0)^{\frac{1}{2}}} u_{1s}(r)\left[3X\left(\frac{1}{r_s}-\frac{1}{r_0}\right)-Xr^2\left(\frac{1}{r_s^3}-\frac{1}{r_0^3}\right)\right]d\tau$$
$$+\int_{r>r_0} \frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{(\Omega_0)^{\frac{1}{2}}} u_{1s}(r)\left[-\frac{2X}{r}+\frac{3X}{r_0}-\frac{Xr^2}{r_0^3}\right]d\tau. \quad (24)$$

The first integral in (24) is

$$3X\left(\frac{1}{r_s}-\frac{1}{r_0}\right)A_{1s}(k)-X\left(\frac{4\pi}{\Omega_0}\right)^{\frac{1}{2}}\left(\frac{1}{r_s^3}-\frac{1}{r_0^3}\right)\int_0^{\infty} r^2\frac{\sin kr}{k}P_{1s}(r)dr. \quad (25)$$

The second integral is small and varies less rapidly with r_s: for $\mathbf{k}=0$ it may be reduced by using (22) to

$$-6X\left(\frac{4\pi}{\Omega_0}\right)^{\frac{1}{2}}\frac{P_{1s}(r_0)}{(-E_{1s})^{\frac{1}{2}}r_0^2}\left[1+\frac{1}{r_0(-E_{1s})^{\frac{1}{2}}}\right]. \quad (26)$$

The F integrals with $\nu \neq \nu'$ are much less important than the other overlap integrals, and may for many purposes be neglected. Finally, when core functions are used which go to zero at $r=r_1$, as described above, the I_{nl} and F integrals with $\nu=\nu'$ can be calculated with any desired precision by expanding $\exp(i\mathbf{k}\cdot\mathbf{r})$ and $(U-V)u_{nl;k'}$ in spherical harmonics; if this is done they reduce to a number of radial integrals to be calculated numerically.

(5) Interpolation in the calculation of $U[\mathbf{K}]$ and $A_{nl}(k)$. When calculations are made for several values of the lattice constant, the integral occurring in (18) is needed for a great many values of K; an integral of similar form is needed also for $A_{nl}(k)$. The values of the integral are most conveniently found by interpolation between directly calculated values for a few values of K. In making interpolations or extrapolations of this sort use can often be made of an analytic function which fits the directly calculated points fairly well and has the correct asymptotic behavior. Such a function can be obtained, for example, by setting a "screened potential"

$$v(r) = -\frac{2X-2Z\exp(-\sigma r)}{r} \text{ in (18).} \quad (27)$$

This gives

$$\int_0^{\infty} r\left[v(r)+\frac{2X}{r}\right]\sin Krdr = -\frac{2ZK}{K^2+\sigma^2}. \quad (28)$$

Similarly if we set $P_{1s}=2b^{\frac{1}{2}}re^{-br}$ (the form for a hydrogenic eigenfunction) we have

$$A_{1s}(k) = \left(\frac{4\pi}{\Omega_0}\right)^{\frac{1}{2}}\frac{4b^{5/2}}{(k^2+b^2)^2}. \quad (29)$$

(6) Simplification for eigenfunctions of certain symmetry types. It often happens that eigenfunctions occur, in or near the occupied portion of the valence electron band, which belong to symmetry types which do not occur in the core bands. For example a number of eigenfunctions φ_k occur for lithium slightly above the Fermi surface, which are odd under the operation of inversion about any nucleus. Since all functions of the corresponding reduced wave vector in the $1s$ band are even under inversion, each such φ_k is orthogonal to the core by virtue of its symmetry alone. Any linear combination of plane waves having the given symmetry type will thus be orthogonal to the true core functions, and also to the approximate core functions constructed from the u_{nlm}. So all we need to do is to set up a secular equation relative to combinations of plane waves belonging to this symmetry type; this secular equation will be the same as if the corresponding linear combination of χ functions had been used instead of plane waves, because the contributions from all terms in (13) beyond the first two, and from all terms in (14) beyond the first, would add up to zero. Thus to calculate energies of eigenfunctions of such symmetry types only the Fourier coefficients $U[\mathbf{K}]$ are needed. In most cases it will suffice to use the second-order Schrödinger perturbation, and it can be shown that the second-order perturbations due to successive

plane wave combinations of the given symmetry will decrease rapidly. This is most easily seen by considering a particular example. Suppose that $\chi_1^- = (2/\Omega)^{\frac{1}{2}} \sin \mathbf{k}\cdot\mathbf{r}$ is a plane wave combination of the given symmetry type (this occurs for some of the lithium eigenfunctions mentioned above). The matrix element of H connecting χ_1^- with $(1/\Omega)^{\frac{1}{2}} \exp[i(\mathbf{k}+\mathbf{K})\cdot\mathbf{r}]$ will be the difference of two Fourier coefficients of the potential; for very large K it will be of order $1/K^3$, since $U[\mathbf{K}]$ is of order $1/K^2$, and even for smaller K's it will be much less than either Fourier coefficient alone. The second-order perturbation series will thus converge extremely rapidly. This in itself is of course no guarantee that this series gives a good approximation to the eigenvalue: if we were to consider functions of the symmetry type of $\chi_1^+ = (2/\Omega)^{\frac{1}{2}} \cos \mathbf{k}\cdot\mathbf{r}$, the second-order perturbation series would converge—less rapidly than above, but still fairly rapidly—to an energy value far below that of the valence electron eigenfunction of the same symmetry type (because χ_1^+ is not orthogonal to the core functions of the 1*s* band). However, it is likely that when we are working with functions of the symmetry type of χ_1^- the off-diagonal matrix elements in the secular determinant will decrease so rapidly to zero that we can set a very small upper bound to the difference between the true eigenvalue and the energy yielded by the second-order perturbations.

In conclusion I wish to thank Professors J. C. Slater and E. P. Wigner for reading and discussing this manuscript.

JUNE 15, 1940 PHYSICAL REVIEW VOLUME 57

Optical and Magnetic Properties of Magnetite Suspensions

Surface Magnetization in Ferromagnetic Crystals

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(Received April 10, 1940)

A magneto-optic effect discovered by W. R. Grove in 1845 is suggested as explaining, in part, at least, peculiarities of ferromagnetic colloid patterns on ferromagnetic crystal surfaces.

C. W. HEAPS[1] has recently investigated the optical transparency of a suspension of magnetite in oil, as modified by magnetic fields applied parallel to the light path or at right angles thereto. He explains the effects as due to changes in the area obstructed by rectilinear rows of particles, magnetically linked, when these are turned from random directions so as to lie parallel with the applied field. A field intensity of only a few oersteds produced nearly maximal effect.

The increase in transparency of a suspension of magnetite in water when a magnetic field is applied parallel to the light path was first demonstrated by W. R. Grove at a social meeting of the Royal Institution, in London, on January 8, 1845. Since the report is short, relatively inaccessible, and ignored for so long a time, it is quoted in full.[2]

"Jan. 8, 1845.—Prof. Grove communicated to the proprietors at this their first *soirée* for the season, some of the leading discoveries in physical science during the past year. Of electrical subjects, M. Matteucci's researches were described; with experimental illustrations; as also the magnetic note. In reference to the latter, Mr. Grove detailed a curious experiment that had occurred to him, and which bore greatly on the subject. A glass tube open at the ends, but protected along its length with a copper jacket, was filled

[1] C. W. Heaps, Phys. Rev. **57**, 528–531 (1940). See also Q. Majorana, Accad. Lincei, Rendiconti [6a] **29**, 11–14 (1939) January; Ricerca Scient. **10**, 783–789 (1939), September.

[2] C. V. Walker, Elec. Mag. **1**, 601 (1845), April.

with water, in which was suspended powdered magnetic oxide of iron. On looking through the tube at distant objects a considerable portion of the light was intercepted by the heterogeneous arrangement of the particles of the oxide; but on passing a current through a coil placed round the tube, these particles assumed a symmetrical character, and much more light was transmitted. This experiment was alluded to as an illustration of the molecular polarization and vibration that is attributed to the particles of an iron rod when a musical note is obtained under the influence of the current."

Grove does not seem to have gone any further, and if interest was aroused it was soon forgotten in the furor which followed Faraday's announcement, to the same group, on November 3, 1845, of the rotation of the plane of polarization of light in traversing a transparent medium parallel to an applied magnetic field.[3] Grove alluded to his own magneto-optic effect twice more,[4] and then gave up.

The magneto-optic effect discovered by Grove and systematized by Heaps may be of value in explaining the relatively opaque bands in a ferromagnetic colloid close to the smooth surface of a ferromagnetic crystal when the normal component of the magnetic field is altered by external means.[5] The shifting of these bands when a considerable normal field is reversed was at first interpreted as a bodily transport of colloid particles from boundaries where local block magnetizations were *vis-à-vis* to boundaries where they were *dos-y-dos*, or vice versa. More recently, Elmore[6] has argued that a sufficiently dilute colloid would not be so concentrated at block boundaries because field gradients, though adequate in magnitude for sufficiently rapid transport, have relatively small components parallel to the surface along which the motions seem to occur. He prefers to explain rapid concentration by invoking interparticle magnetic fields once the particles have been properly aligned by fields of more remote origin. We now suggest that the transparent regions are not actually free from colloid particles but are merely more transparent because, in them, long chains of particles have suitable directions.

The conditions of illumination favor this explanation. The light used comes through the microscope objective, which has a relatively great numerical aperture, so that a divergent magnetic field, with approximate symmetry about a line (or plane) normal to the surface, is well adapted to align chains for easy illumination and easy seeing of a reflecting surface beneath the colloid layer. It will also be noticed that convection currents along the surface would not be observable if the chains in the drifting suspension turned as they were bid by the nonmoving field which they traversed. The fact that drift is actually not as troublesome as it was expected to be therefore favors the present hypothesis.

A test of the idea would seem to require a change in type of illumination, perhaps involving a complete blackout of the underlying metallic surface, and I cannot yet see how to manage this without losing the pattern under observation. A good nonoptical method of estimating particle density, which would also serve, seems equally difficult to devise. I am therefore publishing this note in the hope that someone more actively interested in these matters can devise and conduct a crucial experiment.

[3] The date given is that of a meeting of the Council of the Royal Institution, as reported in the Athenaeum, No. 941, of November 8, 1845, on page 1080. The public presentation, before the Royal Society of London was on November 27, 1845.

[4] First, on May 24, 1849, in presenting a paper on another subject before the Royal Society of London. This was briefly abstracted: Phil. Mag. [3] **35** 153–154 (1849); Ann. d. Physik [2] **78**, 567–568 (1849); Proc. Roy. Soc. London, **5**, 826–827 (1851). Second, in 1850, in the second edition of his *Correlation of Physical Forces*, on pages 72–73.

[5] L. W. McKeehan and W. C. Elmore, Phys. Rev. **46**, 226–228 (1934).

[6] W. C. Elmore, Phys. Rev. **51**, 982–988 (1937). Data on the magnetic properties of colloid particles are to be found in his more recent paper, Phys. Rev. **54**, 1092–1095 (1938).

LETTERS TO THE EDITOR

Prompt publication of brief reports of important discoveries in physics may be secured by addressing them to this department. Closing dates for this department are, for the first issue of the month, the eighteenth of the preceding month, for the second issue, the third of the month. Because of the late closing dates for the section no proof can be shown to authors. The Board of Editors does not hold itself responsible for the opinions expressed by the correspondents.

Communications should not in general exceed 600 words in length.

Artificial Radioactivity Induced in Zr and Mo*

The radioactivity produced in Zr and Mo is so complicated that we have not finished the study of it. Although some of the early data have been published,[1] we think it worth while to report the results so far obtained, because in the fission products some of these radioactive isotopes have been found[2] and some others are also expected among them.

The main results obtained on each chemically separated fraction are shown in Tables I and II.

Cb^{95} (1 h) and Cb^{*93} (55 d) are produced by the β-decay of Zr^{95} (17 h) and $Zr,^{93}$ (63 d), respectively. The former was independently found by Grosse and Booth to have 75 min. Because of the very low energy β-rays (the range distribution studied by the cloud chamber looks like a line spectrum), Nb (55 d) is supposed to be a genetically related isomer of Nb^{93}.

Ma^{101} (9 m) and Ma^{99} (6 h) are produced by the β-decay of Mo^{101} (19 m) and Mo^{99} (64 h), respectively as have been reported already.[3] A search for Mo^{89} and Nb^{91} has been

TABLE I. *Radioactivity induced in Zr. The fast neutrons are from the* Li+D *reaction.*

BOMBARDMENT	OBSERVED PERIODS														
Slow n no chem.	6m ±1	—	18m ±3	1h ±0.2	—	—	—	—	17h ±1	—	—	—	—	—	63d ±10
Slow n Cb frac.	—	—	—	1h ±0.2	—	—	—	—	—	—	—	—	—	long ~55d	—
Slow n Zr. frac.	—	—	—	—	—	—	—	—	17h ±1	—	—	—	—	—	long ~63d
Fast n no chem.	—	10m ±1	—	—	—	—	—	3h ±1	—	66h ±2	68h ±2	70h ±10	—	long ~55d	long ~63d
Fast n Cb frac.	—	—	—	—	—	—	—	—	—	—	—	—	—	long ~55d	—
Fast n Zr frac.	—	—	—	—	—	—	—	3h ±0.5	—	—	68h ±2	70h ±5	—	—	63d ±5
Fast n Y frac.	—	—	—	—	—	—	—	—	—	66h ±2	—	—	—	—	—
Fast n Sr frac.	—	—	—	—	—	2.5h ±0.3	—	—	—	—	—	—	long (55d?)	—	—
D no chem.	—	—	—	—	90m ±3	—	2.5h ±0.3	—	—	3d ±0.2	—	—	—	55d ±5	63d ±5
Sign	β^-	β^+	β^-	β^-	β^-	β^-	β^-	β^-	β^-	β^-	β^+	β^-	β^-	β^-	β^-
Reactions	(n, γ)	—	?	Zr^{95} β^- decay	?	(n, α)	—	(n, p)	(n, γ)	(n, p) (d, α)	$(n, 2n)$	?	(n, α)	Zr^{93} β^- decay	(n, γ) (d, p)
K-U Upper Limit Cloud Chamber	—	—	—	—	—	—	—	—	1.17	2.11	1.05 0.57	1.17	—	Line ~0.20	0.48 0.20
Upper Limit Mev Absorption	~1.9	—	—	—	~1.5	0.5	—	~2.0	1.2	—	1.0	—	1.9	~0.15	~0.25
Assignment	Zr^{97} ?	N^{13} Imp.	?	Cb^{95}	?	Sr^{*87}	Mn^{56} Imp.	Y^{88} ?	Zr^{95}	Y^{90}	Zr^{89}	?	Sr^{89}	Cb^{*93}	Zr^{93}
Periods	6m ±1	—	18m ±3	62m ±5	90m ±3	2.5h ±0.3	—	3h ±0.5	17h ±1	66h ±2	68h ±2	70h ±5	—	55d ±5	63d ±5
Class	*B*	—	*D*	*A*	*D*	*B*	—	*C*	*A*	*A*	*A*	*C*	*C*	*B*	*A*
Remarks Intensity	Prot. *W*	Imp. *VS*	*W*	*W*	*S*	*W*	Imp. *W*	*W*	Prot. *VS*	*VS*	*VS*	*W*	*VW*	*S*	*S*

TABLE II. *Radioactivity induced in* Mo. *The fast neutrons are from the* Li+D *reaction.*

BOMBARDMENT	OBSERVED PERIODS										
Slow n Ma frac.	9m ±1	—	—	—	6h ±1	—	—	—	—	—	—
Slow n Mo frac.	—	—	19m ±1	—	—	—	64h ±1	—	—	—	—
Slow n Cb+Zr	—	—	—	1h ±0.2	—	~17h	—	?	~11d	—	long
Fast n Ma frac.	—	—	—	—	6h ±1	—	—	—	—	—	—
Fast n Mo frac.	—	17m ±1	—	—	—	—	64h ±1	—	—	—	—
Fast n Cb franc.	—	—	—	62m ±5	—	—	—	—	11d ±2	long (55d?)	—
Fast n Zr frac.	—	—	—	—	—	17h ±1	—	68h ±2	—	—	long (63d?)
Sign Reactions	β^- Mo^{101} β^- decay	β^+ $(n, 2n)$	β^- (n, γ)	β^- (n, p) Zr^{95} β^- decay	β^- Mo^{99} β^- decay	β^- (n, α)	β^- (n, γ) $(n, 2n)$	β^+ (n, α)	β^- (n, p)	Nb^{93} β^- decay	β^- (n, α)
Remarks Intensity	*S*	*VS*	prot. *VS*	*S*	*W*	*S*	*VS*	*S*	*W*	*VW*	*VW*
K-U Upper Limit Cloud Chamber	1.14	2.65	1.78	—	—	—	1.44 1.01	—	—	—	—
Upper Limit Mev Absorption	1.2	—	1.9	1.8	soft	1.2	—	1.0	—	soft	soft
Assignment	Ma^{101} (3)	Mo^{91}	Mo^{101} (3)	Cb^{95}	Ma^{99} (3)	Zr^{95}	Mo^{99}	Zr^{89}	Cb^{92} (4)	Cb^{*93}	Zr^{93}
Periods	9m ±1	17m ±1	19m ±1	62m ±5	6h ±1	17h ±1	64h ±1	68h ±2	11d ±1	55d ±5	63d ±5
Class	*A*	*A*	*A*	*B*	*B*	*B*	*A*	*B*	*B*	*C*	*C*

made in vain, and it is suspected for the existence of a stable nucleus Nb^{91}.

A more detailed report will be published shortly in the *Proceedings of the Physico Mathematical Society of Japan*.

* These results were reported at the general meeting of the Phys. Math. Soc. Japan held on April 3, 1940.

[1] Sagane, Kojima, Miyamoto and Ikawa, Phys. Rev. **54**, 970 (1938).

[2] Hahn and Strassmann, Naturwiss. **27**, 451 (1939); Grosse and Booth, Phys. Rev. **57**, 665 (1940).

[3] Seaborg and Segrè, Phys. Rev. **55**, 808 (1939); Sagane, Kojima, Miyamoto and Ikawa, Phys. Rev. **57**, 751 (1940).

[4] Sagane, Kojima, Miyamoto and Ikawa, Proc. Phys. Math. Soc. Japan **22**, 174 (1940).

[5] The installation of the cyclotron has been assisted by the Japan Society for the Promotion of Scientific Research, Mitsui Ho-Onkai Foundation, Tokyo Electric Light Company, Japan Wireless Telegraph Company and K. Hattori and Company.

The authors are very grateful to Dr. Y. Nishina and Professor K. Kimura for their valuable help and discussions. The experiment has been aided by grants given by Oji Paper Manufacturing Company and the Department of Education of the Government as the expenditure for the Scientific Researches.

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On the Equilibrium of the Components of Cosmic Radiation at Sea Level

To examine the behavior of the equilibrium between the two components of the cosmic radiation, i.e., the soft component (absorbed at sea level by 10 cm of lead) and the hard component, absorption measurements have been taken up to 10 cm of lead at different zenith inclinations. The counter telescope consisted of three pairs of counters of 4.3×30 cm² (1 mm brass walls) filled with argon and alcohol, and arranged as per Fig. 1. The layer of lead was in position *P*. For computation of the side showers, the two central counters were moved outside of the alignment as shown in Fig. 1 with dotted lines.

The experiments were first carried out in the inside of the Institute in front of a large window and afterwards in a wood cabin in the garden. The measurements taken in such different surroundings gave the same results. Table I refers to the measurements taken in the Institute with the exception of $\theta = 0°$. The results are also shown in Fig. 2.

Contrary to what is generally believed, it appears that the equilibrium between the hard and soft components is not reached at sea level, but only under a layer equivalent to about 12 m of H_2O, if the minimum of the curve of Fig. 2 must be interpreted in this way.

Definite conclusions cannot be obtained for the soft and hard equilibria because the angular scattering in the multi-

TABLE I. *Observations in cosmic-ray absorption at different zenith inclinations.*

	$P=0$ cm Pb			$P=10$ cm Pb		
INCLIN.	COINC./MIN.	SHOWERS/MIN.	C_0	COINC./MIN.	SHOWERS/MIN.	C_{10}
0°	$\frac{5688}{893}=6.38$	$\frac{184}{401}=0.46$	5.92 ±0.08	$\frac{8075}{1675}=4.83$	$\frac{115}{582}=0.2$	4.63 ±0.05
20°	$\frac{16415}{2807}=5.85$	$\frac{730}{1636}=0.45$	5.40 ±0.05	$\frac{11819}{2533}=4.67$	$\frac{425}{2865}=0.15$	4.52 ±0.05
30°	$\frac{14435}{3003}=4.81$	$\frac{958}{2569}=0.37$	4.44 ±0.04	$\frac{11725}{2935}=4.00$	$\frac{448}{3067}=0.15$	3.85 ±0.04
45°	$\frac{5064}{1574}=3.22$	$\frac{484}{1795}=0.27$	2.95 ±0.045	$\frac{5519}{2249}=2.45$	$\frac{282}{2037}=0.14$	2.31 ±0.04
60°	$\frac{8542}{5380}=1.59$	$\frac{378}{2231}=0.17$	1.42 ±0.02	$\frac{4410}{3514}=1.255$	$\frac{244}{1956}=0.12$	1.13 ±0.02
75.5°	$\frac{3491}{6771}=0.515$	$\frac{513}{4379}=0.117$	0.398±0.007	$\frac{2956}{8021}=0.368$	$\frac{390}{4788}=0.081$	0.287±0.007

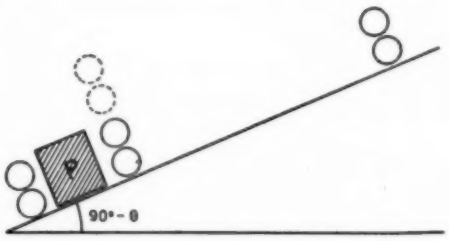

FIG. 1. Arrangement of counters.

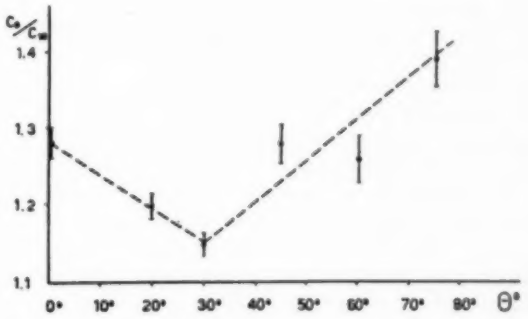

FIG. 2. Ratio for different zenith inclinations of the cosmic-ray intensity without lead absorption and with 10 cm of lead.

plicative theory is not yet known with exactness. On the other hand, we think that we can state that the scattering causes the increase in the percentage of the soft component at larger inclinations. It should be pointed out that in our measurements, the side showers were taken into consideration. In our case the correction of the showers is not very much, but it is certainly not to be neglected. Since this correction was not generally carried out before in measurements of this kind, the results obtained up to date do not lend themselves very well to comparison; in less suitable geometrical arrangements the showers can completely change the measurements.

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The Force on the Cathode of an Arc

Some years ago several papers[1] were published on the subject of the force exerted on the cathode of an arc. This phenomenon is interesting especially at low gas pressures because the force is then larger. Tanberg has explained this force by the reaction of a high velocity vapor stream emerging from the cathode. Although this explanation presumes very high speeds of the vapor stream no better explanation has been given.[2] Robertson[3] has again taken up this question and has investigated especially the variation of the force as a function of the gas pressure without giving an explanation of this dependence. The purpose of this note is to show that the variation with the gas pressure can also be explained by the high velocity stream.

If the density of the gas is high the high velocity stream is slowed down by giving its momentum to the gas. As this momentum is directed away from the cathode the transfer of it to the gas results in a diminishing of the gas pressure on the cathode and therefore partially compensates the force exerted on the cathode by the high velocity stream. If the gas pressure is low and the force exerted by it on the front surface of the cathode is much smaller than the reaction of the high velocity stream, the diminishing of the gas pressure can have no measurable effect. If, however, the gas pressure increases, the reduction of it in front of the cathode by the transfer of the momentum will be increased. This would explain the decrease of the total force on the cathode with increasing gas pressure measured by Robertson. On account of the uncertainty of the area of the cathode which must be taken into con-

sideration and of the ratio of the momentum transfer, the calculation of the decrease of the force is practically impossible. We can compute only whether the measured decrease of the total force *can* be caused by a decrease of the gas pressure.

For this purpose we shall take the forces at 15 amp. measured by Tanberg in vacuum and by Robertson at 1 mm Hg for a copper arc indicated in Fig. 4 of Robertson's paper. We assume that the transfer of the momentum of the stream to the gas diminishes the gas pressure to one-half. The measured forces in vacuum and at 1 mm Hg are 270 dynes and 160 dynes, respectively. The reduction is 110 dynes and the reduced gas pressure must then be effective on 0.168 cm². It naturally must be not larger than the area of the face of the cathode spot but may be larger than the area of the cathode spot. The face of the cathode in the experiments of Robertson had an area of 0.316 cm². The area of the cathode spot is 0.00105 cm² at 15 amperes if we calculate it with the ratio of 0.007 mm² amp. measured in vacuum by Tanberg and Berkey. At 1 mm Hg the current density is rather higher and the area of the cathode spot therefore smaller. We see that the calculated area lies between the two limits. The explanation seems therefore to be possible.

Lamar[4] also has pointed out that the absence of the force on the cathode in an arc at higher gas pressures may be due to the compensating effect of holding back the gas pressure from the cathode spot. But, according to his opinion this effect occurs as the result of convection currents set up around the cathode if this is of relatively small dimensions and therefore can not occur if the cathode has a large surface area.

Now, if my explanation of the effect is right the compensating effect is not confined to small cathode areas because the reduction of the gas pressure in front of the cathode spot and its near surroundings does not augment the gas pressure on areas of the cathode which lie further away from the cathode spot and because the high velocity stream is only slowed down by the gas and is not reflected back to the cathode. On the contrary for the full development of the compensating effect the area of the cathode must be larger than a certain minimum.

In conclusion I should like to thank Dr. F. Luedi for his suggestion to explain the results obtained by Robertson.

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January 18, 1940.

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Induced β-Activity of Uranium by Fast Neutrons

In the course of experiments on the fission of uranium by fast neutrons,[1] besides fission products the uranium fraction showed a β-activity with a period 6.5 days.

This activity was induced appreciably only by fast neutrons obtained by bombarding lithium with 3-Mev. deuterons from our cyclotron. The experimental procedure was as follows.

A few grams of uranium oxide, U_3O_8, carefully purified and freed from its disintegration products were exposed to fast neutrons for more than fifty hours. After the exposure, a uranium fraction (U_3O_8) was separated and purified from all possible elements produced by fission as well as from its own disintegration products. The most care was given to the removal of lanthanum from the sample, the procedure taking as long as one day. The activity of the irradiated uranium was compared with that of a nonirradiated sample, in order to subtract the growing β-activity due to disintegration products of uranium. The difference thus obtained shows a 6.5-day period. This activity is probably due to U^{237} produced from U^{238} through loss of a neutron, as in the case of the production of UY from thorium.[2] If this is the case, we have here a member of the missing radioactive family $4n+1$.

The sign of the β-rays was shown to be negative and consequently we suspected the production of a radioactive element of atomic number 93, the chemical properties of which are probably homologous to rhenium. From the decay curve it is clear that its period must be very long, if it exists. To search for such an element, the irradiated uranium oxide, which was freed from fission products as well as its own disintegration products as above mentioned, was left for about 7 days, and was then dissolved in nitric acid. The solution, after an addition of perrhenic acid, was treated with ammonium sulphide and then acidified with sulphuric acid. The precipitated rhenium sulphide, after the removal of contaminated sulphur by carbon bisulphide, was examined for β- and α-activities. Neither of them could be found within the error of our experiments. We may thus conclude, as in the case of 23-minute uranium,[3] that the 6.5-day uranium decays also into a very long-lived 93 element. The detailed accounts of the experiments will be given elsewhere.

The above investigations were carried out as a part of the work of the Atomic Nucleus Sub-Committee of the Japan Society for the Promotion of Scientific Research. We acknowledge the assistances given by our laboratory colleagues in connection with the irradiation of samples and by Messrs. N. Saito and N. Matuura regarding the chemical separations.

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An Absorption Law for Total Radiation Measurements

Beer's law, which may be stated in the form

$$I_\lambda = I_{0\lambda} e^{-kL}$$

applies to the transmission of radiant energy by homogeneous media or solutions, for specific wave-lengths or for narrow spectral intervals. Forsythe[1] indicates a weighted-mean absorptivity for total radiation which assumes knowledge of both $I_0(\lambda)$ and $k(\lambda)$.

Schack[2] suggests the linear relation $k=a+b\lambda$ and obtains for the band transmission

$$I=\int_\lambda^{\lambda+\Delta\lambda} I_{0\lambda} e^{-(a+b\lambda)L} d\lambda = I_M \Delta\lambda \frac{1-e^{-KL}}{KL}$$

(integrated for the special case $k=0$ at λ and $k=K$ at $\lambda+\Delta\lambda$, an unnecessary restriction).

The symbols in the foregoing are defined as follows: $I_\lambda d\lambda$ = transmitted intensity for an interval $d\lambda$ at wave-length λ. $I_{0\lambda} d\lambda$ = incident intensity for an interval $d\lambda$ at wave-length λ. $I_M \Delta\lambda$ = incident intensity for the band $\Delta\lambda$. k = absorption coefficient at wave-length λ. L = path length (replaced by pL for an atmospheric path over which pressure, but not composition, varies).

Schack proceeds to a more accurate representation of $k(\lambda)$ by imagining the spectral lines and fringes within the band to be rearranged in order of their absorption coefficients, whereupon the ascending function $k(\lambda)$ may be well represented by a series of straight lines $k=a_n+b_n\lambda$, with intersections at $k_1, k_2, \cdots k_{n-1}$, the extreme values being k_0 and k_n. The band transmission is now expressed by the sum of n integrals, resulting as follows:

$$I = I_M \Delta\lambda \sum_1^n \frac{x_n}{(k_n-k_{n-1})L}(e^{-k_{n-1}L}-e^{-k_nL}).$$

Here $\Sigma x_n = 1$, the x values being fractions of the incident energy to which the straight-line k values $a_n+b_n\lambda$ apply.

In setting up empirical equations to express the variation of atmospheric absorption, or emissivity, as a function of path length (from thermopile measurements covering the entire infra-red spectrum) it is more convenient and equally permissible to imagine the total spectral incident energy subdivided without regard to wave-length but in descending order of absorption coefficient, i.e., from lines of near opacity to spectral regions in which the atmosphere is practically transparent.

If we now consider I and I_0 to be intensities of total infra-red radiation (transmitted and incident, respectively) the incident intensity I_0 may be conveniently thought of as divided into blocks (for example, three in number) within which the decrease of k is approximately linear, as illustrated in Fig. 1.

The fractions x_1, x_2, and x_3, of course, total unity, as in Schack's treatment, but all pretense of knowledge of $k(\lambda)$ is abandoned. Instead we express $k(x)=a_n+b_nx$ in each "block" of energy.

The total spectral transmissivity becomes

$$\frac{I}{I_0}=\int_0^1 e^{-Lk(x)}dx,$$

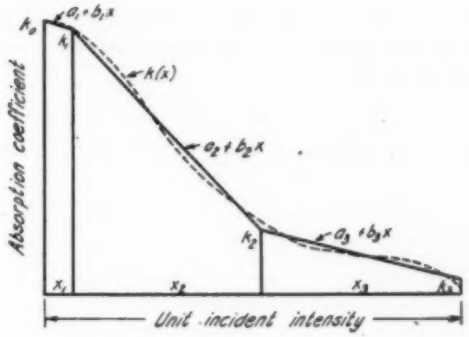

FIG. 1.

which is closely approximated by

$$\frac{I}{I_0}=\int_0^{x_1} e^{-(a_1+b_1x)L}dx+\int_{x_1}^{x_1+x_2} e^{-(a_2+b_2x)L}dx+\int_{x_1+x_2}^1 e^{-(a_3+b_3x)L}dx$$

$$=\frac{x_1}{(k_0-k_1)L}(e^{-k_1L}-e^{-k_0L})+\frac{x_2}{(k_1-k_2)L}(e^{-k_2L}-e^{-k_1L})$$

$$+\frac{x_3}{(k_2-k_3)L}(e^{-k_3L}-e^{-k_2L}).$$

It may be shown that the transmission terms reduce to their respective x values for $L=0$, giving the necessary total transmissivity of unity for zero path length. Moreover, it is obvious that for increasing L all terms approach zero.

Another form of empirical expression for band transmissivity consists of a sum of terms $x_ne^{-k_nL}$, which corresponds to a step-function representation of $k(x)$. In general this form will require more terms for satisfactory agreement with the data, particularly where wide spectral ranges are covered.

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Eigenvalue Problem of the Dirac Electron

Let $m_0c^2=\epsilon_0$ be the rest energy and $\tau_0=c/a$ be the rest signal time of an electronic particle whose Dirac[1] signal radius is $a=\gamma e^2/m_0c^2$ with a factor γ whose most probable value is $\frac{2}{3}$ or $2\pi\cdot\frac{2}{3}$. A combination of quantum theory and relativity, based on ideas of Born[2] and the writer,[3] has led to the result that the product of ϵ_0 and τ_0, instead of being of the order of h, has the surprisingly small value

$$\epsilon_0\tau_0=\hbar\mu=\hbar\cdot 0.0299, \qquad (1)$$

where μ is the smallest root of the transcendental equation $2\pi\mu[\psi_0(\mu)]^2=1$, and is related to Sommerfeld's constant, $\alpha=\mu/\gamma$. If γ were $2\pi\cdot\frac{2}{3}$ then our μ would yield $\alpha=1/140$. However, γ cannot be determined uniquely as long as one deals with a single particle; a signal requires at least two particles. Furthermore, our result holds for the point electron only.

The eigenvalue problem of the Dirac spin electron has to be a generalization of the former scalar problem which had led to the integral equation:

$$(\mu/2\pi)^3 \int\int S_P(R|R')\psi(R')dV_0'dW_0=\psi(R). \quad (2)$$

Here dV_0' and dW_0 are invariant elements in space and in momentum space, and

$$S_P(R|R')=\exp\{i\mu(P\cdot R-R'-E\cdot T-T')\}$$
$$E=\pm(P^2+1)^{\frac{1}{2}},\quad T=\pm(R^2+1)^{\frac{1}{2}},\quad P=p/m_0c,\quad R=r/a. \quad (3)$$

The only change for the electron with spin is that S is now a matrix, and that there are four ψ-functions. Using the spinor $\psi_+\psi_-$ and its spin-conjugate $\chi_+{}^+\chi_-{}^+$ and calling them $\psi_1\psi_2\psi_3\psi_4$ we arrive at the following set of integral equations as a generalization of (2):

$$(\mu/2\pi)^3 \int\int dV_0'dW_0 S_P(R|R') \times \begin{Bmatrix} (E+P_z)\psi_3(R)+(P_x-iP_y)\psi_4(R') \\ (P_x+iP_y)\psi_3(R')+(E-P_z)\psi_4(R') \\ (E-P_z)\psi_1(R')-(P_x-iP_y)\psi_2(R') \\ -(P_x+iP_y)\psi_1(R')+(E+P_z)\psi_2(R') \end{Bmatrix} = \begin{matrix} \psi_1(R) \\ \psi_2(R) \\ \psi_3(R) \\ \psi_4(R). \end{matrix} \quad (4)$$

For small P these equations reduce to (2). The bracket has exactly the same structure as the Dirac's differential equations except that in the latter $|E|$ stands for the operator $(\hbar/im_0c^2)\partial/\partial t$, etc. In contrast to the case of the point electron, we are far from knowing the exact eigenfunctions and eigenvalues. Nevertheless it seems interesting that the spin leads to a new eigenvalue problem (although there is no external electromagnatic field) simply because of the presence of large momenta in the probability distribution.

The physical meaning of our signal equation $(ct)^2-r^2=a^2$ is shown by its application (Born): Whereas in a *stationary* state of momentum p the probability amplitude of "transition" from the point r at the time t, to the point r' at the same time t, is $S_P(r|r')=\psi(r)\psi^*(r')=\exp\{2i\pi(p\cdot r-r')\}$, Born assumed that the transition probability in a *stationary* state from r at the time t to r' at the advanced or retarded time t' is given by (3).

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The Spin Angular Momenta of Elementary Particles

In the present state of the field theories it is not clear to what extent a consistent formulation of the "particle" as distinguished from the "field" aspects of the formalism can be made. The most characteristic condition in the electron theory is that the square of any component of the spin is an absolute invariant, but this is possible only for spin 0 and $\frac{1}{2}$. I have investigated the looser condition that the squared magnitude of the space part of the spin is a Lorentz invariant. This condition insures that the "magnitude" of the spin is the same for all Lorentz systems, and so has an invariant significance. This gives the three relations

$$S_{42}S_{12}+S_{43}S_{13}-i\hbar S_{41}=0 \quad \text{(cyclic)}, \quad (1)$$

which lead at once to

$$S \equiv S_{12}{}^2+S_{23}{}^2+S_{31}{}^2=S_{14}{}^2+S_{24}{}^2+S_{34}{}^2. \quad (2)$$

If we assume the spin expressible in terms of an operator 4-vector by

$$S_{kl}=\lambda[\beta_k, \beta_l],$$

the general commutation relations become

$$(\beta_k\beta_l\beta_m+\beta_m\beta_l\beta_k)-(\beta_l\beta_k\beta_m+\beta_m\beta_k\beta_l) = (i\hbar/\lambda)(\delta_{km}\beta_l-\delta_{lm}\beta_k), \quad (3)$$

which are satisfied by both the Dirac ($\lambda=-i\hbar/4$) and the Duffin[1] ($\lambda=-i\hbar$) operators.

With

$$\beta^2=\beta_1{}^2+\beta_2{}^2+\beta_3{}^2$$
$$S^2=\lambda^2\{\beta^2(\tfrac{1}{2}\beta^2+i\hbar/\lambda)-\tfrac{1}{2}(\beta_1\beta^2\beta_1+\beta_2\beta^2\beta_2+\beta_3\beta^2\beta_3)\}. \quad (4)$$

For the Duffin operators satisfying

$$\beta_k\beta_l\beta_m+\beta_m\beta_l\beta_k=\beta_k\delta_{lm}+\beta_m\delta_{lk} \quad (5)$$

we get

$$S^2=(i\hbar)^2\beta^2(\beta^2-3).$$

Introducing condition (1) there results

$$(i\hbar)^2(\beta_1\beta_4+\beta_4\beta_1)(\beta_2{}^2+\beta_3{}^2-1)=0. \quad \text{(cyclic)}$$

It is not apparent that these conditions are in conflict with Eqs. (5), but from the explicit matrices given by Kemmer[2] for the irreducible representations it is found that they are satisfied only for the trivial case of zero spin.

From the general representation $D_{k,l}$ of the Lorentz group[3] obtained by separation of the spin tensor into two self-dual tensors $\mathbf{\Gamma}$ and $\mathbf{\Delta}$ as in Dirac's method,[4] where

$$\mathbf{\Gamma}\times\mathbf{\Gamma}=i\hbar\mathbf{\Gamma},\quad \mathbf{\Delta}\times\mathbf{\Delta}=i\hbar\mathbf{\Delta},\quad [\Gamma_k, \Delta_l]=0$$

conditions (1) become

$$\mathbf{\Gamma}\times\mathbf{\Delta}=0,$$

yielding

$$\mathbf{S}^2=\mathbf{\Gamma}^2+\mathbf{\Delta}^2.$$

By simultaneous quantization of these three operators we get

$$s(s+1)=k(k+1)+l(l+1),$$

where s, k, and l are all to be integral or half-integral. This is possible only for $l=0$, $k=s$ or $k=0$, $l=s$. The wave function in terms of spinors has components of the form

$$\varphi^{\lambda\mu\ldots}\qquad \chi_{\sigma'\epsilon'\ldots},$$

each with $2s$ indices. Since the momentum operator spinor has one primed and one unprimed index, it is not feasible to connect these spinors by a linear wave equation except for $s=\frac{1}{2}$. The possibility of using families of secondary functions along the lines discussed by Fierz[5] has not yet been investigated.

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Anomalous Dispersion and Absorption of Electric Waves in Solutions of Amino-Acids and Dipeptides

The dielectric constants and absorption coefficients of aqueous solutions of several amino-acids and dipeptides, listed in Table I, have been measured for electric waves of $\lambda=25.5$ cm ($\nu=1.18\times10^9$ cycles). The method used was a modification of Drude's second method. The waves were generated as described earlier.[1] The table shows the results obtained: $\Delta\epsilon'/c$ is the so-called dielectric increment per mole and $\Delta\epsilon''/c$ is the absorption coefficient increment per mole; ϵ' and ϵ'' being the real and the imaginary parts of the dielectric constant $\epsilon=\epsilon'-i\epsilon''$. The increments $\Delta\epsilon_0/c$ of the static dielectric constant per mole were taken from the literature.[2,3] The table shows also the relaxation times τ calculated from our data for the dielectric increment by the use of Debye's theory formulae with the Wyman's formula $\tau=a+bp$ in place of the Clausius-Mossoti relation (p—volume polarization; a and b—Wyman's constants). The relaxation times τ_0 calculated for spherical molecules from molecular volumes are given for comparison in the last column.

Attention may be drawn to the following points: (a) the anomalous dispersion and absorption were increasing with increasing molecular size; (b) the dielectric increment was independent of concentration within the limits of error; (c) the absorption coefficient increment tended to increase with the concentration; this could be quantitatively accounted for as being due to the change in relaxation time with the change in viscosity to which ϵ'' is very sensitive when frequency corresponds to the edge of the dispersive region.

As is well known the observed static dielectric increments of simple straight-chain dipolar ions in a polar solvent have been accounted for statistically on the assumption of free rotation about the various single bonds of the chain, irrespective of whether polarization in a static field is accomplished by deformation or by orientation of the entire molecule.[4,5] At high frequencies, however, the dielectric properties of the solution should depend upon the mechanism of polarization, each mode of deformation or orientation having its own relaxation time. The behavior of the higher peptides of glycine strongly suggests polarization by deformation of a flexible chain, since no anomalous dispersion has been observed at frequencies corresponding to the rotation of the molecules as a whole.[3] The present data for the simple dipeptides, on the other hand, suggest orientation of these molecules, and are further consistent with the assumption of an asymmetry factor of about 2. The amino-acids, with the very doubtful exception of glycine, also give evidence of relaxation times not greatly different from those corresponding to molecular rotation. A fuller discussion of the experiments will be published elsewhere.

TABLE I. *Dielectric constants, absorption coefficients, and relaxation times.*

SUBSTANCE	$\Delta\epsilon_0/c$	CONCENTRATION RANGE	$\Delta\epsilon'/c$	$\Delta\epsilon''/c$	$\tau\times10^{11}$	$\tau_0\times10^{11}$
Glycine	22.8	0.2–2.0	21.3	14.3	2.60	6.61
α-Alanine	23.3	0.4–0.8	17.4	13.8	6.55	8.50
β-Alanine	34.8	0.4–0.8	25.7	19.1	6.72	8.50
Glycylglycine	71.1	0.2–0.4	28.7	41.2	14.94	10.82
Glycylalanine	72.3	0.2–0.4	20.2	39.5	20.28	12.71
Alanylglycine	71.5	0.2	18.9	40.3	21.28	12.71

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Radioactive Element 93

Last year a nonrecoiling 2.3-day period was discovered[1] in uranium activated with neutrons, and an attempt[2] was made to identify it chemically, leading to the conclusion that it is a rare earth. Impressed by the difficulties raised by this identification, the authors independently decided that the subject was worth further investigation. In Berkeley it was found that: (1) If a layer of $(NH_4)_2U_2O_7$ with about 0.1 mm air equivalent stopping power, placed in contact with a collodion film of 2 mm air equivalent, is activated by neutrons from the cyclotron, the 2.3-day period appears strongly in the uranium layer, and not at all in the collodion, which shows a decay curve parallel to, and 1/7 as strong as, that of a paper "fission catcher" behind it. One day after bombardment the uranium layer has five times the activity of the fission catcher. This shows that the 2.3-day period has a range of <0.1 mm air and an intensity larger than all the long period fission products together. (2) When a thin layer of uranium is bombarded with and without cadmium around it, the fission product intensity is changed by a large factor, while the 2.3-day period and the 23-minute uranium period are only slightly changed, and their ratio remains constant. Also absorption of resonance neutrons by uranium changes these two periods in the same ratio, suggesting a genetic relation between them, and the consequent identification of the longer period with element 93. In Washington it was found that the 2.3-day period probably does not behave consistently as a rare earth, since attempts to concentrate it chemically with the rare earths from activated uranium failed, although it is known to have an intensity large compared with that of the rare earth fission products.

At this stage of the investigation one of the authors (P.H.A.) came to Berkeley on a visit, and a combined attack was made. With pure 2.3-day substance from thin uranium layers, the chemical properties were investigated, and a very characteristic difference from the rare earths was soon found; namely, the substance does not precipitate with HF in the presence of an oxidizing agent (bromate in strong acid). In the presence of a reducing agent (SO_2) it precipitates quantitatively with HF. Cerium was used as a carrier. This property explains the erratic nature of previous chemical experiments in which the oxidizing power of the solution was not controlled. Further chemical

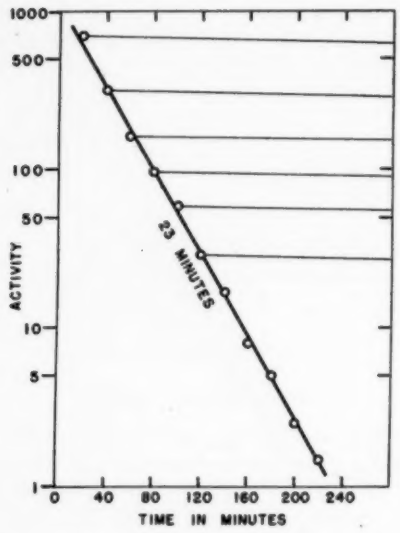

FIG. 1. Growth of 2.3-day 93^{239} from 23-minute U^{239}. The points indicate the activities of successive fluoride extractions, plotted at the times of extraction. Decay measurements were made a day later on the first six fractions, and the resulting slopes are shown on the plot.

experiments showed that in the reduced state with a thorium carrier it precipitates with iodate, and in the oxidized state with uranium as sodium uranyl acetate. It also precipitates with thorium on the addition of H_2O_2. It precipitates in basic solution if carbonate is carefully excluded. These properties indicate that the two valence states are very similar to those of uranium (U^{++++} and UO_2^{++} or $U_2O_7^{--}$), the chief difference from that element being in the value of the oxidation potential between the two valences, such that the lower state is more stable in the new element. It is interesting to note that the new element has little if any resemblance to its homolog rhenium; for it does not precipitate with H_2S in acid solution, is not reduced to the metal by zinc in acid solution, and does not have an oxide volatile at red heat. This fact, together with the apparent similarity to uranium, suggests that there may be a second "rare earth" group of similar elements starting with uranium.

The final proof that the 2.3-day substance is the daughter of the 23-minute uranium is the demonstration of its growth from the latter. For this experiment activated uranium was purified twice by precipitation as sodium uranyl acetate, which was dissolved in HF and saturated with SO_2. Then equal quantities of cerium were added at twenty-minute intervals and the precipitates filtered out. The first precipitate, made immediately after purification, carried all the fluoride-precipitable contaminations and was discarded; its weakness indicated a very good purification. The activities of the others are plotted in Fig. 1.

A preliminary study of the radiations from 93^{239} shows that it emits continuous negative beta-particles with an upper limit of 0.47 Mev, and a weak complex spectrum of low energy gamma-rays (<0.3 Mev) and probably x-rays. The question of the behavior of its daughter product 94^{239} immediately arises. Our first thought was that it should go to actinouranium by emitting an alpha-particle. We sought for these by preparing a strong sample (11 millicuries) of purified 93 and placing it near a linear amplifier in a magnetic field to deflect the beta-particles. From this experiment we conclude that, if alpha-particles are emitted, their half-life must be of the order of a million years or more; the same experiment showed that if spontaneous fission occurs its half-life must be even greater. We wish to express our gratitude to the Rockefeller Foundation and the Research Corporation, whose financial support made this work possible.

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[1] E. McMillan, Phys. Rev. **55**, 510 (1939).
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Masses of Stable Nuclei from Ne^{20} to Fe^{57}

The observation of the proton groups in (dp) and (αp) reactions recently made in this laboratory, together with a knowledge of beta-ray maximum energies makes it possible to give reasonably correct values for isotopic masses as far as Fe^{57} with a few exceptions. These values are given in Table I. The values from Ne^{22} to A^{38} are derived entirely from transmutation data, and mass-spectrograph values are given in parentheses for comparison where they are available. Above K^{39} the accuracy is less and reliance has to be placed on mass-spectrograph measurements of Ti^{48}, Fe^{56} [1] and Cr^{52}.[2]

Such mass values are useful in checking theories of nuclear structure and to facilitate a bird's-eye view of the trend of mass values a quantity called here the "mass deviation" is plotted against the mass number. This is simply the deviation from a whole number plotted on a sloping scale which enables the whole range of elements to be plotted in a small space. Precisely defined it is $M_A - A + 0.001(A-20)$ where M_A is the mass of an element of

TABLE I. *Isotopic masses.*

Ne^{20}	19.99881 (Bainbridge)	A^{40}	39.97504 (Bainbridge)
Ne^{21}	21.00018 (20.99968)	Ca^{40}	39.9745
Ne^{22}	21.99864 (Bainbridge)	K^{41}	40.9739
Na^{23}	22.99680	Ca^{42}	41.9711
Mg^{24}	23.99189	Ca^{43}	42.9723
Mg^{25}	24.99277	Ca^{44}	
Mg^{26}	25.99062	Sc^{45}	44.9701
Al^{27}	26.98960	Ti^{46}	45.9678
Si^{28}	27.98639 (27.9866)	Ti^{47}	
Si^{29}	28.98685 (28.9866)	Ti^{48}	47.9651 (Dempster)
Si^{30}	29.98294	Ti^{49}	48.9664
P^{31}	30.98457 (30.9843)	Ti^{50}	49.9632 (Dempster)*
S^{32}	31.98306 (31.9823)	V^{51}	50.9587
S^{33}	32.98260	Cr^{52}	51.9575 (Aston)
S^{34}	33.97974	Cr^{53}	52.9572
Cl^{35}	34.98107	Cr^{54}	
A^{36}	35.97852	Fe^{54}	53.9601 (Dempster)*
Cl^{37}	36.97829 (36.9779)	Mn^{55}	54.9653
A^{38}	37.97544	Fe^{56}	55.9608 (Dempster)
K^{39}	38.97518	Fe^{57}	56.9609

* Considerably less accurate than the other values given.

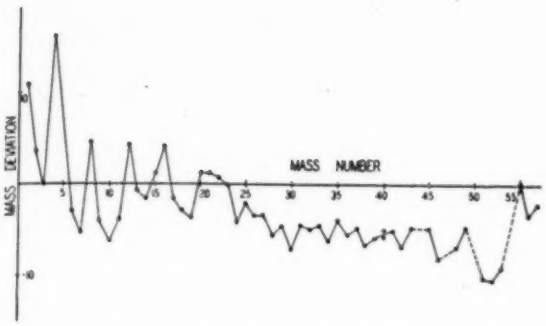

FIG. 1. "Mass deviation" as a function of mass number.

mass number A. The well-known variations up to Ne^{20} are shown and the absence of any marked irregularities from Ne^{20} to Ca^{40} is in good agreement with the supposition that the $3d$ shell is being filled in this region.[3] After mass number 40 the irregularities become more pronounced and between Cr^{53} and Mn^{55} a strong fluctuation occurs. This strong fluctuation may in part be due to error in the mass of Fe^{56} upon which the points at 55, 56 and 57 depend, but it is doubtful if all the sharp rise can be explained in this way. The indication is that a sudden increase in mass occurs upon the addition of either the 30th neutron or the 25th proton. Since if short range forces operate we expect the $4f$ shell to be in process of being filled for either of these numbers, it is hard to understand the presence of a fluctuation at all. A possible explanation is that the fluctuation corresponds to a simultaneous filling of both proton and neutron shells. The simplest way of achieving this is to suppose that after Ca^{40} the neutrons fill the $3p$ and $3s$ shells while the protons occupy $3s$ and $4s$ shells. This would then give a simultaneous filling of shells at 30 neutrons and 24 protons, meaning a low mass for Cr^{54} and a relatively high value for Mn^{55}. A measurement of the packing fractions of the three chromium isotopes would be of great interest in checking this suggestion.

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June 1, 1940.

[1] A. J. Dempster, Phys. Rev. **53**, 64 (1938).
[2] F. W. Aston, Nature **141**, 1096 (1938).
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Anomalous Scattering of Neutrons by Helium

It is known[1,2] that neutrons of about one Mev energy show an anomalously high scattering cross section in helium. The variation of the scattering cross section with neutron energy in the region from zero up to one Mev has not been carefully investigated, and it was the purpose of the present work to obtain data in that region.

Neutrons from the reaction $C^{12}+D^2 \rightarrow N^{13}+n'+Q$ were used as a source. Magnetically analyzed deuteron beams up to 1.6 Mev energy were produced by the Rice pressure Van de Graaff generator. Since $Q=-0.28$ Mev, neutrons in the forward direction have an energy of 1.2 Mev at this maximum bombarding energy. The paraffin target was not more than about 15 kv in thickness.

The ionization current produced in a Wulf electroscope filled with 45 pounds of hydrogen was observed when the neutrons from the carbon reaction were allowed to enter the instrument within about 20° of the forward direction of the incident deuteron beam. The ionization current was obtained as a function of the incident deuteron energy. The corresponding energy of the neutrons was calculated, using the above Q value. The spread in neutron energy arising from target penetration of the deuterons, angular deviation from the forward direction (0°–20°), and spread of deuteron energy is not more than about 3 percent.

The electroscope was then filled with 25 pounds of helium and the observations were repeated, the ionization current in helium being obtained as a function of neutron energy.

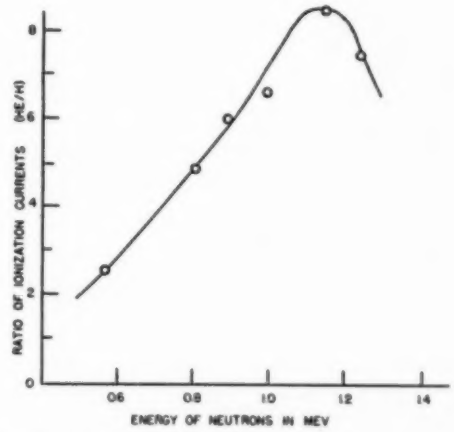

FIG. 1. Ratio of observed ionization currents in helium and hydrogen as a function of neutron energy. The maximum has been normalized to 8.6, the value of σ_{He}/σ_H for neutrons of about one Mev energy.

It is known[3] that strong γ-rays are obtained when carbon is bombarded by deuterons, and the ionization current produced by these γ-rays was taken into account. This, however, together with the natural background, constituted only a fraction of the total ionization current observed.

The ratios of these corrected ionization currents in helium and hydrogen are shown in Fig. 1. The ratio plotted in Fig. 1 may also be regarded as a measure of the ratio σ_{He}/σ_H of the cross section for neutron scattering in helium and in hydrogen, if (a) the recoil protons and recoil α-particles show the same spatial distribution, and (b) the ratio of the number of ions collected in helium to the number collected in hydrogen remains constant for various neutron energies. Since both recoil protons and recoil α-particles are believed to show spherical symmetry in their spatial distribution in a moving coordinate system, condition (a) is probably satisfied. The electroscope collected ions over a region large compared to any recoil track lengths, and hence condition (b) is also fulfilled.

The ratio σ_{He}/σ_H has been normalized in Fig. 1 to 8.6 at its maximum value; this is the previously observed[1,2] ratio of the absolute cross sections at that point. At the time of this experiment it was not possible to obtain neutrons of higher energy from carbon because of generator voltage limitations, but it is hoped that these measurements may be extended in the near future. Below 0.6 Mev neutron energy, the yield is quite small, so that ionization currents in the electroscope were of the order of the natural background. Furthermore, small effects from $d-d$ neutrons, arising from deuteron contamination of the target, would become correspondingly quite large at these low energies, and hence points below 0.6 Mev were not thought to be reliable. However, Carroll and Dunning[4] have shown that σ_{He}/σ_H for thermal neutrons is only 0.05.

The curve rises smoothly to a maximum at about 1.1 Mev; if it is continued symmetrically beyond this maximum, the resonance shows a half-width of nearly 0.6 Mev. This is in agreement with cloud-chamber work which has been done here.[2]

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The Rice Institute,
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May 15, 1940.

[1] H. Staub and W. E. Stephens, Phys. Rev. **55**, 131 (1939).
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Dispersion of Supersonic Waves in Cylindrical Rods

In a recent issue of this journal a paper[1] appeared on the dispersion of supersonic waves in cylindrical rods. A number of experimental results were reported and compared with the theory of Giebe and Blechschmidt.[2] It was shown that agreement was good out to the region where the theoretical "dead zone" begins, but at higher frequencies the theory quoted was quite inadequate.

As certain work, which was reported by the writer some years ago, seems to have escaped attention, it seems worth while to recall here some of the results obtained at that time which are relevant to the recent paper by Shear and Focke and which amplify certain of the conclusions reached by these authors.

When a sound wave is propagated down a cylindrical tube filled with liquid, it has been shown[3-5] that dispersion of the wave occurs at frequencies corresponding to those of the resonant radial vibrations of the column. In the neighborhood of these resonant frequencies the longitudinal wave is propagated only with difficulty and what might be called a "dead zone" exists.

That this "dead zone" also exists for tubes was shown later by Giebe and Blechschmidt,[2] who obtained satisfactory agreement between their experimental results and data calculated from the simple theory of coupled oscillations.

It was thought at one time[6] that this same type of anomalous dispersion occurred for solid cylindrical rods. The curves upon which these views were based, however, were apparently inaccurate at the higher frequencies and it seems probable that certain resonances were attributed to longitudinal vibrations which actually belonged to other modes, e.g., torsional or flexural. Later experimental work by Röhrich[7] and now confirmed by Shear and Focke[1] has shown that the "dead zone" does not exist for solid rods, and, in fact, it is not predicted by the theory which should correctly be applied to this particular case. This theory is that due originally to Pochhammer,[8] summarized by Love,[9] and discussed in detail a few years ago in several papers.[10,6]

In a concluding paper published in 1934[11] the writer considered longitudinal vibrations for the three cases mentioned above and offered a suggestion for the absence in the last case of the "dead zone" which was so striking a phenomenon in the first two cases. This suggestion was that for the solid rod shear forces were of very great importance and would prevent anomalous dispersion and hence the dead zone from occurring. A theoretical discussion showed this to be the case.

Thus it appears that the limitations of the theory discussed by Shear and Focke exist because this particular theory is applicable only to the special and limited case of a tube. If in each case the appropriate theory be considered, it has been shown that the experimental confirmation is quite gratifying.

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National Research Council Laboratories,
Ottawa, Canada,
May 30, 1940.

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[3] G. S. Field, Can. J. Research **5**, 131 (1931).
[4] R. W. Boyle, D. K. Froman and G. S. Field, Can. J. Research **6**, 102 (1932).
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[7] K. Röhrich, Zeits. f. Physik **73**, 813 (1932).
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On the Dynamics of Complex Fission

In a letter of the same title[1] we pointed out that our coefficients of the higher order terms in the potential energy of distortion of a liquid drop, the radius of which is described by the expression $r=R(1+a_0+a_2P_2+a_4P_4)$, do not check those given by Bohr and Wheeler.[2] Dr. Wheeler has explained that the discrepancy arises from the fact that the coefficients α_2 and α_4 as used in their Eq. (22) are not as defined in their paper and that (22) is obtained by using another definition based on spheroidal harmonics.[3] Our definition gives a_4 positive at the a_2-a_4 saddle point for the range of x from zero to one. The interpretation of this fact as given in Section two of our letter is partially incorrect. The equatorial bulge which a positive a_4 would introduce is so small that it does not compensate for the equatorial constriction automatically brought in by the second harmonic, as was pointed out to us by Dr. Wheeler. For values of x sufficiently far from unity the drop in its critical shape develops a concavity about the equatorial belt. We cannot conclude that the positive value for a_4 is

in itself an indication of ternary or asymmetric binary fission. The possibility of ternary fission has not been sufficiently investigated.

We should state again that the coupling terms between odd and even harmonics provide an explanation of the marked asymmetry of fission. In the calculations so far there has been found evidence for two modes of fission. One is by way of a saddle point for which the drop when in the critical shape has a symmetrical form ($a_3=0$). The other is by way of a twofold saddle point for which the critical shape is not symmetrical ($a_3\neq 0$). Which mode requires the less activation energy cannot be decided at the present stage of the calculations. The odd-even coupling terms will have an effect in determining the final products of either mode. It must be emphasized that higher order terms, which are being introduced, may materially affect the entire situation.

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May 16, 1940.

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[2] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
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Neutron-Induced Radioactivity of Tungsten

On bombarding tungsten with slow neutrons, Fermi and his co-workers[1] found an activity decaying with a period of about 1 day, and McLennan and others[2] and Jaeckel[3] obtained, as its half-life, 23 hours and 24.5±1 hours, respectively. They ascribed this activity to the reaction $W^n(n,\gamma)W^{n+1}$, but the isotope concerned was not determined.

To see if there are some other periods, pure metallic tungsten powder (99.97 percent purity) was irradiated for about 100 hours (intermittently) with neutrons produced in the Tokyo cyclotron according to the following conditions:

	Target	Sample
fast neutrons:	Li+D, enclosed in Cd+B box, immediately behind a Li-target.	
slow neutrons:	Li+D, enclosed in a paraffin block, about 30 cm apart from the target.	
	Be+D, enclosed in a paraffin block.	

A Lauritsen type electroscope was used to observe the activity. In all of these samples, besides the well-known 24-hour tungsten period (in our case 24.0±0.1 hours), a long-lived activity of half-life 77±3 days was found.

Chemical separation showed that the carrier of this long-lived activity is also tungsten and is neither Ta nor Hf. With a thin-walled G-M counter and magnetic field, it was found that both carriers emit negative electrons.

From the relative intensities of both periods in the above three cases, it was found that the shorter period is produced practically only with slow neutrons and the longer one both with fast and slow neutrons. The above results lead to the conclusion that 24-hour activity is due to W^{187} and 77-day activity to W^{185}.

By using the Al absorption method and the range-energy relation,[4] the upper limits of energy of the β-rays were measured and were found to be 1.1±0.1 Mev (24 hours), and about 0.4−0.5 Mev (77 days), respectively. Both periods were accompanied by a considerable amount of γ-rays. A more detailed report will be published shortly in the *Scientific Papers of the Institute of Physical and Chemical Research.*

In conclusion the writer wishes to express his best thanks to Professor H. Nagaoka for his interest and kind encouragement throughout this work, and to Dr. Y. Nishina for his kind suggestions and discussions, and to Mr. T. Shirai for assistance in chemical separations of the samples.

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JUN 26 1940

THE PHYSICAL REVIEW

VOLUME 57 *Second Series* NUMBER 12

JUNE 15, 1940

Published for the

AMERICAN PHYSICAL SOCIETY

by the

AMERICAN INSTITUTE OF PHYSICS
Incorporated

LANCASTER, PA., AND NEW YORK, N. Y.

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Subscription Price

United States and Possessions and Canada $15.00
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Back Numbers

Complete set: Series 2,[1] Vol. 1, 1913–Vol. 56, 1939—$425.00
Yearly rate: $16.50
Single copies: $1.50 each prior to July, 1929; $.75 each after July, 1929.

Subscriptions and orders for back numbers should be addressed to Prince and Lemon Streets, Lancaster, Pennsylvania, or to the American Institute of Physics, 175 Fifth Avenue, New York, New York.

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The Physical Review is published semi-monthly at Prince and Lemon Streets, Lancaster, Pennsylvania.

Entered at the Post Office at Lancaster, Pennsylvania, as second class matter.

Acceptance for mailing at special rate of postage provided for in section 1103, Act of October 3, 1917, authorized September 6, 1935.

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